

VERTRAG ÜBER DIE INTERNATIONALE ZUSAMMENARBEIT AUF DEM GEBIET DES PATENTWESENS

PCT

INTERNATIONALER RECHERCHENBERICHT

(Artikel 18 sowie Regeln 43 und 44 PCT)

Aktenzeichen des Anmelders oder Anwalts pctS3	WEITERES VORGEHEN	
Internationales Aktenzeichen PCT/AT 00/ 00167	Internationales Anmeldedatum (Tag/Monat/Jahr) 21/07/2000	siehe Mitteilung über die Übermittlung des internationalen Recherchenberichts (Formblatt PCT/ISA/220) sowie, soweit zutreffend, nachstehender Punkt 5 (Frühestes) Prioritätsdatum (Tag/Monat/Jahr) 21/06/1999
Anmelder SCHRÖDL MANFRED		

Dieser internationale Recherchenbericht wurde von der Internationalen Recherchenbehörde erstellt und wird dem Anmelder gemäß Artikel 18 übermittelt. Eine Kopie wird dem Internationalen Büro übermittelt.

Dieser internationale Recherchenbericht umfaßt insgesamt 2 Blätter.



Darüber hinaus liegt ihm jeweils eine Kopie der in diesem Bericht genannten Unterlagen zum Stand der Technik bei.

1. Grundlage des Berichts

- a. Hinsichtlich der **Sprache** ist die internationale Recherche auf der Grundlage der internationalen Anmeldung in der Sprache durchgeführt worden, in der sie eingereicht wurde, sofern unter diesem Punkt nichts anderes angegeben ist.
- ☐ Die internationale Recherche ist auf der Grundlage einer bei der Behörde eingereichten Übersetzung der internationalen Anmeldung (Regel 23.1 b)) durchgeführt worden.
- b. Hinsichtlich der in der internationalen Anmeldung offenbarten **Nucleotid- und/oder Aminosäuresequenz** ist die internationale Recherche auf der Grundlage des Sequenzprotokolls durchgeführt worden, das
- ☐ in der internationalen Anmeldung in schriftlicher Form enthalten ist.
- ☐ zusammen mit der internationalen Anmeldung in computerlesbarer Form eingereicht worden ist.
- ☐ bei der Behörde nachträglich in schriftlicher Form eingereicht worden ist.
- ☐ bei der Behörde nachträglich in computerlesbarer Form eingereicht worden ist.
- ☐ Die Erklärung, daß das nachträglich eingereichte schriftliche Sequenzprotokoll nicht über den Offenbarungsgehalt der internationalen Anmeldung im Anmeldezeitpunkt hinausgeht, wurde vorgelegt.
- ☐ Die Erklärung, daß die in computerlesbarer Form erfaßten Informationen dem schriftlichen Sequenzprotokoll entsprechen, wurde vorgelegt.

2. ☐ Bestimmte Ansprüche haben sich als nicht recherchierbar erwiesen (siehe Feld I).
3. ☐ Mangelnde Einheitlichkeit der Erfindung (siehe Feld II).

4. Hinsichtlich der Bezeichnung der Erfindung

- ☒ wird der vom Anmelder eingereichte Wortlaut genehmigt.
- ☐ wurde der Wortlaut von der Behörde wie folgt festgesetzt:

5. Hinsichtlich der Zusammenfassung

- ☒ wird der vom Anmelder eingereichte Wortlaut genehmigt.
- ☐ wurde der Wortlaut nach Regel 38.2b) in der in Feld III angegebenen Fassung von der Behörde festgesetzt. Der Anmelder kann der Behörde innerhalb eines Monats nach dem Datum der Absendung dieses internationalen Recherchenberichts eine Stellungnahme vorlegen.

6. Folgende Abbildung der **Zeichnungen** ist mit der Zusammenfassung zu veröffentlichen: Abb. Nr. 2 ☐ keine der Abb.

- ☒ wie vom Anmelder vorgeschlagen
- ☐ weil der Anmelder selbst keine Abbildung vorgeschlagen hat.
- ☐ weil diese Abbildung die Erfindung besser kennzeichnet.

INTERNATIONALER RECHERCHENBERICHT

nationales Aktenzeichen
PCT/AT 00/00167

A. KLASSIFIZIERUNG DES ANMELDUNGSGEGENSTANDES
IPK 7 H02K16/00 H02K16/02

Nach der Internationalen Patentklassifikation (IPK) oder nach der nationalen Klassifikation und der IPK

B. RECHERCHIERTE GEBIETE

Recherchierter Mindestprüfstoff (Klassifikationssystem und Klassifikationssymbole)
IPK 7 H02K

Recherchierte aber nicht zum Mindestprüfstoff gehörende Veröffentlichungen, soweit diese unter die recherchierten Gebiete fallen

Während der internationalen Recherche konsultierte elektronische Datenbank (Name der Datenbank und evtl. verwendete Suchbegriffe)
EPO-Internal, WPI Data, PAJ

C. ALS WESENTLICH ANGESEHENE UNTERLAGEN

Kategorie*	Bezeichnung der Veröffentlichung, soweit erforderlich unter Angabe der in Betracht kommenden Teile	Betr. Anspruch Nr.
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A	EP 0 769 403 A (TOYOTA MOTOR CO LTD) 23. April 1997 (1997-04-23) Absatz '0006! Spalte 8, Zeile 1 - Zeile 41; Abbildungen 1,2	1-3,6-8, 11
A	EP 0 725 474 A (NIPPON DENSO CO) 7. August 1996 (1996-08-07) Spalte 4, Zeile 12 - Zeile 20 Spalte 4, Zeile 44 - Zeile 51 Spalte 5, Zeile 15 - Zeile 32 Spalte 6, Zeile 30 - Zeile 33; Abbildung 1	1-3,6-9
A	EP 0 800 951 A (TOYOTA MOTOR CO LTD) 15. Oktober 1997 (1997-10-15) Zusammenfassung, Anspruch 1 Abbildung 1	1-3,6-8

☐ Weitere Veröffentlichungen sind der Fortsetzung von Feld C zu entnehmen

☒ Siehe Anhang Patentfamilie

- * Besondere Kategorien von angegebenen Veröffentlichungen:
- *A* Veröffentlichung, die den allgemeinen Stand der Technik definiert, aber nicht als besonders bedeutsam anzusehen ist
- *E* älteres Dokument, das jedoch erst am oder nach dem internationalen Anmeldedatum veröffentlicht worden ist
- *L* Veröffentlichung, die geeignet ist, einen Prioritätsanspruch zweifelhaft erscheinen zu lassen, oder durch die das Veröffentlichungsdatum einer anderen im Recherchenbericht genannten Veröffentlichung belegt werden soll oder die aus einem anderen besonderen Grund angegeben ist (wie ausgeführt)
- *O* Veröffentlichung, die sich auf eine mündliche Offenbarung, eine Benutzung, eine Ausstellung oder andere Maßnahmen bezieht
- *P* Veröffentlichung, die vor dem internationalen Anmeldedatum, aber nach dem beanspruchten Prioritätsdatum veröffentlicht worden ist

- *T* Spätere Veröffentlichung, die nach dem internationalen Anmeldedatum oder dem Prioritätsdatum veröffentlicht worden ist und mit der Anmeldung nicht kollidiert, sondern nur zum Verständnis des der Erfindung zugrundeliegenden Prinzips oder der ihr zugrundeliegenden Theorie angegeben ist
- *X* Veröffentlichung von besonderer Bedeutung; die beanspruchte Erfindung kann allein aufgrund dieser Veröffentlichung nicht als neu oder auf erfinderischer Tätigkeit beruhend betrachtet werden
- *Y* Veröffentlichung von besonderer Bedeutung; die beanspruchte Erfindung kann nicht als auf erfinderischer Tätigkeit beruhend betrachtet werden, wenn die Veröffentlichung mit einer oder mehreren anderen Veröffentlichungen dieser Kategorie in Verbindung gebracht wird und diese Verbindung für einen Fachmann naheliegend ist
- *G* Veröffentlichung, die Mitglied derselben Patentfamilie ist

Datum des Abschlusses der internationalen Recherche

9. November 2000

Absendedatum des internationalen Recherchenberichts

16/11/2000

Name und Postanschrift der Internationalen Recherchenbehörde
Europäisches Patentamt, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax: (+31-70) 340-3016

Bevollmächtigter Bediensteter

Zoukas, E

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
PCT/AT 00/00167

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP 0769403 A	23-04-1997	JP 9170533 A	30-06-1997
		DE 69608200 D	15-06-2000
		US 5934395 A	10-08-1999
EP 0725474 A	07-08-1996	JP 3052786 B	19-06-2000
		JP 8340663 A	24-12-1996
		JP 3052820 B	19-06-2000
		JP 9056010 A	25-02-1997
		US 5744895 A	28-04-1998
		US 5917248 A	29-06-1999
		CN 1141859 A	05-02-1997
		JP 9056126 A	25-02-1997
EP 0800951 A	15-10-1997	JP 3003573 B	31-01-2000
		JP 9266601 A	07-10-1997
		US 5973460 A	26-10-1999

Angaben zu Veröffentlichungen, die zur selben Patentfamilie gehören

PCT/AT 00/00167

Im Recherchenbericht angeführtes Patentdokument	Datum der Veröffentlichung	Mitglied(er) der Patentfamilie	Datum der Veröffentlichung
EP 0769403 A	23-04-1997	JP 9170533 A	30-06-1997
		DE 69608200 D	15-06-2000
		US 5934395 A	10-08-1999
EP 0725474 A	07-08-1996	JP 3052786 B	19-06-2000
		JP 8340663 A	24-12-1996
		JP 3052820 B	19-06-2000
		JP 9056010 A	25-02-1997
		US 5744895 A	28-04-1998
		US 5917248 A	29-06-1999
		CN 1141859 A	05-02-1997
		JP 9056126 A	25-02-1997
EP 0800951 A	15-10-1997	JP 3003573 B	31-01-2000
		JP 9266601 A	07-10-1997
		US 5973460 A	26-10-1999

PATENT COOPERATION TREATY

PCT

COMMUNICATION IN CASES FOR WHICH
NO OTHER FORM IS APPLICABLE

From the INTERNATIONAL BUREAU

To:

KRAUSE, Peter
Sagerbachgasse 7
A-2500 Baden
AUTRICHE

Date of mailing (day/month/year) 21 November 2000 (21.11.00)	REPLY DUE see paragraph 1 below
Applicant's or agent's file reference pctS3	
International application No. PCT/AT00/00167	International filing date (day/month/year) 21 June 2000 (21.06.00)
Applicant SCHRÖDL, Manfred	

1. ☐ REPLY DUE within _____ months/days from the above date of mailing
- ☐ NO REPLY DUE, however, see below
- ☒ IMPORTANT COMMUNICATION
- ☐ INFORMATION ONLY

2. COMMUNICATION:

Following a communication from the receiving Office (RO/AT), the International Bureau confirms that the International Filing Date of the present international application has been corrected to read:

21 June 2000 (21.06.00)

instead of 21 July 2000 (21.07.00) .

A copy of this communication is being sent to the receiving Office, the International Searching Authority and to the designated Offices that have already been informed of their designation.

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland Facsimile No. (41-22) 740.14.35	Authorized officer Ellen Moyse Telephone No. (41-22) 338.83.38
--	--

PATENT COOPERATION TREATY



PCT

COMMUNICATION IN CASES FOR WHICH
NO OTHER FORM IS APPLICABLE

From the INTERNATIONAL BUREAU

To:

KRAUSE, Peter
Sagerbachgasse 7
A-2500 Baden
AUTRICHE

Date of mailing (day/month/year) 21 November 2000 (21.11.00)	REPLY DUE see paragraph 1 below
Applicant's or agent's file reference pctS3	
International application No. PCT/AT00/00167	International filing date (day/month/year) 21 June 2000 (21.06.00)
Applicant SCHRÖDL, Manfred	

1. ☐ REPLY DUE within _____ months/days from the above date of mailing
- ☐ NO REPLY DUE, however, see below
- ☒ IMPORTANT COMMUNICATION
- ☐ INFORMATION ONLY

2. COMMUNICATION:

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21 June 2000 (21.06.00)

instead of 21 July 2000 (21.07.00) .

A copy of this communication is being sent to the receiving Office, the International Searching Authority and to the designated Offices that have already been informed of their designation.

The International Bureau of WIPO
34, chemin des Colombettes
1211 Geneva 20, Switzerland

Authorized officer

Ellen Moyse

Telephone No. (41-22) 338 83 38

PATENT COOPERATION TREATY

PCT

NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

Commissioner
 US Department of Commerce
 United States Patent and Trademark
 Office, PCT
 2011 South Clark Place Room
 CP2/5C24
 Arlington, VA 22202
 ETATS-UNIS D'AMERIQUE
 in its capacity as elected Office

Date of mailing (day/month/year) 12 March 2001 (12.03.01)	Applicant's or agent's file reference pctS3
International application No. PCT/AT00/00167	Priority date (day/month/year) 21 June 1999 (21.06.99)
International filing date (day/month/year) 21 June 2000 (21.06.00)	
Applicant SCHRÖDL, Manfred	

1. The designated Office is hereby notified of its election made:

☒ in the demand filed with the International Preliminary Examining Authority on:
 15 January 2001 (15.01.01)

☐ in a notice effecting later election filed with the International Bureau on:

2. The election ☒ was
☐ was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO
 34, chemin des Colombettes
 1211 Geneva 20, Switzerland

Facsimile No.: (41-22) 740 14 35

Authorized officer

Henrik Nyberg

Telephone No.: (41-22) 338.83.38

EINGEGANGEN **PCT**

13. Juli 2000

Erl. **ANTRAG**

Der Unterzeichnete beantragt, daß die vorliegende internationale Anmeldung nach dem Vertrag über die internationale Zusammenarbeit auf dem Gebiet des Patentwesens behandelt wird.

Vom Anmeldeamt auszufüllen	
Internationales Aktenzeichen	PCT/AT 2000/167
Internationales Anmeldedatum	21. Juni 2000
Österreichisches Patentamt CSANDE Einlauf- u. Abgangsstelle Name des Anmeldeamts und "PCT, International Application"	
Aktenzeichen des Anmelders oder Anwalts (falls gewünscht) (max. 12 Zeichen) pctS3	

Feld Nr. I BEZEICHNUNG DER ERFINDUNG

Elektrische Maschine

Feld Nr. II ANMELDER

Name und Anschrift: (Familienname, Vorname; bei juristischen Personen vollständige amtliche Bezeichnung. Bei der Anschrift sind die Postleitzahl und der Name des Staats anzugeben. Der in diesem Feld in der Anschrift angegebene Staat ist der Staat des Sitzes oder Wohnsitzes des Anmelders, sofern nachstehend kein Staat des Sitzes oder Wohnsitzes angegeben ist.)

SCHRÖDL Manfred Dipl. Ing. Dr.
 Untere Hauptstrasse 9
 A-7223 Sieggraben
 Österreich

☒ Diese Person ist gleichzeitig Erfinder

Telefonnr.:

Telefaxnr.:

Fernschreibnr.:

Staatsangehörigkeit (Staat):

AT

Sitz oder Wohnsitz (Staat):

AT

Diese Person ist Anmelder für folgende Staaten:

☒ alle Bestimmungsstaaten

☐ alle Bestimmungsstaaten mit Ausnahme der Vereinigten Staaten von Amerika

☐ nur die Vereinigten Staaten von Amerika

☐ die im Zusatzfeld angegebenen Staaten

Feld Nr. III WEITERE ANMELDER UND/ODER (WEITERE) ERFINDER

Name und Anschrift: (Familienname, Vorname; bei juristischen Personen vollständige amtliche Bezeichnung. Bei der Anschrift sind die Postleitzahl und der Name des Staats anzugeben. Der in diesem Feld in der Anschrift angegebene Staat ist der Staat des Sitzes oder Wohnsitzes des Anmelders, sofern nachstehend kein Staat des Sitzes oder Wohnsitzes angegeben ist.)

Diese Person ist:

☐ nur Anmelder

☐ Anmelder und Erfinder

☐ nur Erfinder (Wird dieses Kästchen angekreuzt, so sind die nachstehenden Angaben nicht nötig.)

Staatsangehörigkeit (Staat):

Sitz oder Wohnsitz (Staat):

Diese Person ist Anmelder für folgende Staaten:

☐ alle Bestimmungsstaaten

☐ alle Bestimmungsstaaten mit Ausnahme der Vereinigten Staaten von Amerika

☐ nur die Vereinigten Staaten von Amerika

☐ die im Zusatzfeld angegebenen Staaten

☐ Weitere Anmelder und/oder (weitere) Erfinder sind auf einem Fortsetzungsblatt angegeben.

Feld Nr. IV ANWALT ODER GEMEINSAMER VERTRETER; ODER ZUSTELLANSCHRIFT

Die folgende Person wird hiermit bestellt/ist bestellt worden, um für den (die) Anmelder vor den zuständigen internationalen Behörden in folgender Eigenschaft zu handeln als:

☒ Anwalt

☐ gemeinsamer Vertreter

Name und Anschrift: (Familienname, Vorname; bei juristischen Personen vollständige amtliche Bezeichnung. Bei der Anschrift sind die Postleitzahl und der Name des Staats anzugeben.)

KRAUSE Peter
 Bahnhofstrasse 45
 A-4580 Windischgarsten
 Österreich

Telefonnr.: **++43 2252 477 352**

Telefaxnr.: **++43 2252 477 355**

Fernschreibnr.:

☐ Zustellanschrift: Dieses Kästchen ist anzukreuzen, wenn kein Anwalt oder gemeinsamer Vertreter bestellt ist und statt dessen im obigen Feld eine spezielle Zustellanschrift angegeben ist.

Siehe Anmerkungen zu diesem Antragsformular

Feld Nr. V BESTIMMUNG VON STAATEN

Die folgenden Bestimmungen nach Regel 4.9 Absatz a werden hiermit vorgenommen (bitte die entsprechenden Kästchen ankreuzen; wenigstens ein Kästchen muß angekreuzt werden):

Regionales Patent

- ☒ AP ARIPO-Patent: GH Ghana, GM Gambia, KE Kenia, LS Lesotho, MW Malawi, SD Sudan, SL Sierra Leone, SZ Swasiland, TZ Vereinigte Republik Tansania, UG Uganda, ZW Simbabwe und jeder weitere Staat, der Vertragsstaat des Harare-Protokolls und des PCT ist
- ☒ EA Eurasisches Patent: AM Armenien, AZ Aserbaidschan, BY Belarus, KG Kirgisistan, KZ Kasachstan, MD Republik Moldau, RU Russische Föderation, TJ Tadschikistan, TM Turkmenistan und jeder weitere Staat, der Vertragsstaat des Eurasischen Patentübereinkommens und des PCT ist
- ☒ EP Europäisches Patent: AT Österreich, BE Belgien, CH und LI Schweiz und Liechtenstein, CY Zypern, DE Deutschland, DK Dänemark, ES Spanien, FI Finnland, FR Frankreich, GB Vereinigtes Königreich, GR Griechenland, IE Irland, IT Italien, LU Luxemburg, MC Monaco, NL Niederlande, PT Portugal, SE Schweden und jeder weitere Staat, der Vertragsstaat des Europäischen Patentübereinkommens und des PCT ist
- ☒ OA OAPI-Patent: BF Burkina Faso, BJ Benin, CF Zentralafrikanische Republik, CG Kongo, CI Côte d'Ivoire, CM Kamerun, GA Gabun, GN Guinea, GW Guinea-Bissau, ML Mali, MR Mauretanien, NE Niger, SN Senegal, TD Tschad, TG Togo und jeder weitere Staat, der Vertragsstaat der OAPI und des PCT ist (falls eine andere Schutzrechtsart oder ein sonstiges Verfahren gewünscht wird, bitte auf der gepunkteten Linie angeben)

Nationales Patent (falls eine andere Schutzrechtsart oder ein sonstiges Verfahren gewünscht wird, bitte auf der gepunkteten Linie angeben):

- | | |
|--|--|
| <input checked="" type="checkbox"/> AE Vereinigte Arabische Emirate | <input checked="" type="checkbox"/> LR Liberia |
| <input checked="" type="checkbox"/> AL Albanien | <input checked="" type="checkbox"/> LS Lesotho |
| <input checked="" type="checkbox"/> AM Armenien | <input checked="" type="checkbox"/> LT Litauen |
| <input type="checkbox"/> AT Österreich | <input checked="" type="checkbox"/> LU Luxemburg |
| <input checked="" type="checkbox"/> AU Australien | <input checked="" type="checkbox"/> LV Lettland |
| <input checked="" type="checkbox"/> AZ Aserbaidschan | <input checked="" type="checkbox"/> MA Marokko |
| <input checked="" type="checkbox"/> BA Bosnien-Herzegowina | <input checked="" type="checkbox"/> MD Republik Moldau |
| <input checked="" type="checkbox"/> BB Barbados | <input checked="" type="checkbox"/> MG Madagaskar |
| <input checked="" type="checkbox"/> BG Bulgarien | <input checked="" type="checkbox"/> MK Die ehemalige jugoslawische Republik Mazedonien |
| <input checked="" type="checkbox"/> BR Brasilien | <input checked="" type="checkbox"/> MN Mongolei |
| <input checked="" type="checkbox"/> BY Belarus | <input checked="" type="checkbox"/> MW Malawi |
| <input checked="" type="checkbox"/> CA Kanada | <input checked="" type="checkbox"/> MX Mexiko |
| <input checked="" type="checkbox"/> CH und LI Schweiz und Liechtenstein | <input checked="" type="checkbox"/> NO Norwegen |
| <input checked="" type="checkbox"/> CN China | <input checked="" type="checkbox"/> NZ Neuseeland |
| <input checked="" type="checkbox"/> CR Costa Rica | <input checked="" type="checkbox"/> PL Polen |
| <input checked="" type="checkbox"/> CU Kuba | <input checked="" type="checkbox"/> PT Portugal |
| <input checked="" type="checkbox"/> CZ Tschechische Republik und Gebrauchsmuster | <input checked="" type="checkbox"/> RO Rumänien |
| <input checked="" type="checkbox"/> DE Deutschland und Gebrauchsmuster | <input checked="" type="checkbox"/> RU Russische Föderation |
| <input checked="" type="checkbox"/> DK Dänemark und Gebrauchsmuster | <input checked="" type="checkbox"/> SD Sudan |
| <input checked="" type="checkbox"/> DM Dominica | <input checked="" type="checkbox"/> SE Schweden |
| <input checked="" type="checkbox"/> EE Estland und Gebrauchsmuster | <input checked="" type="checkbox"/> SG Singapur |
| <input checked="" type="checkbox"/> ES Spanien | <input checked="" type="checkbox"/> SI Slowenien |
| <input checked="" type="checkbox"/> FI Finnland und Gebrauchsmuster | <input checked="" type="checkbox"/> SK Slowakei und Gebrauchsmuster |
| <input checked="" type="checkbox"/> GB Vereinigtes Königreich | <input checked="" type="checkbox"/> SL Sierra Leone |
| <input checked="" type="checkbox"/> GD Grenada | <input checked="" type="checkbox"/> TJ Tadschikistan |
| <input checked="" type="checkbox"/> GE Georgien | <input checked="" type="checkbox"/> TM Turkmenistan |
| <input checked="" type="checkbox"/> GH Ghana | <input checked="" type="checkbox"/> TR Türkei |
| <input checked="" type="checkbox"/> GM Gambia | <input checked="" type="checkbox"/> TT Trinidad und Tobago |
| <input checked="" type="checkbox"/> HR Kroatien | <input checked="" type="checkbox"/> TZ Vereinigte Republik Tansania |
| <input checked="" type="checkbox"/> HU Ungarn | <input checked="" type="checkbox"/> UA Ukraine |
| <input checked="" type="checkbox"/> ID Indonesien | <input checked="" type="checkbox"/> UG Uganda |
| <input checked="" type="checkbox"/> IL Israel | <input checked="" type="checkbox"/> US Vereinigte Staaten von Amerika |
| <input checked="" type="checkbox"/> IN Indien | <input checked="" type="checkbox"/> UZ Usbekistan |
| <input checked="" type="checkbox"/> IS Island | <input checked="" type="checkbox"/> VN Vietnam |
| <input checked="" type="checkbox"/> JP Japan | <input checked="" type="checkbox"/> YU Jugoslawien |
| <input checked="" type="checkbox"/> KE Kenia | <input checked="" type="checkbox"/> ZA Südafrika |
| <input checked="" type="checkbox"/> KG Kirgisistan | <input checked="" type="checkbox"/> ZW Simbabwe |
| <input checked="" type="checkbox"/> KP Demokratische Volksrepublik Korea | |
| <input checked="" type="checkbox"/> KR Republik Korea | |
| <input checked="" type="checkbox"/> KZ Kasachstan | |
| <input checked="" type="checkbox"/> LC Saint Lucia | |
| <input checked="" type="checkbox"/> LK Sri Lanka | |

Kästchen für die Bestimmung von Staaten, die dem PCT nach der Veröffentlichung dieses Formblatts beigetreten sind:
☒ Algerien, Mozambik, Belize, Antigua, Barbuda,
☒ Dom. Republik,
 Erklärung bzgl. vorsorglicher Bestimmungen: Zusätzlich zu den oben genannten Bestimmungen nimmt der Anmelder nach Regel 4.9 Absatz b auch alle anderen nach dem PCT zulässigen Bestimmungen vor mit Ausnahme der im Zusatzfeld genannten Bestimmungen, die von dieser Erklärung ausgenommen sind. Der Anmelder erklärt, daß diese zusätzlichen Bestimmungen unter dem Vorbehalt einer Bestätigung stehen und jede zusätzliche Bestimmung, die vor Ablauf von 15 Monaten ab dem Prioritätsdatum nicht bestätigt wurde, nach Ablauf dieser Frist als vom Anmelder zurückgenommen gilt. (Die Bestätigung (einschließlich der Gebühren) muß beim Anmeldeamt innerhalb der Frist von 15 Monaten eingehen.)

Feld Nr. VI PRIORITÄTSANSPRUCH		<input type="checkbox"/> Weitere Prioritätsansprüche sind im Zusatzfeld angegeben.		
Anmeldedatum der früheren Anmeldung (Tag/Monat/Jahr)	Aktenzeichen der früheren Anmeldung	Ist die frühere Anmeldung eine:		
		ationale Anmeldung: Staat	regionale Anmeldung: regionales Amt	internationale Anmeldung: Anmeldeamt
Zeile (1) 21. Juni 1999 21.6.1999	öA 1081/99	AT		
Zeile (2) 15. Dezember 1999 15.12.1999	öA 2115/99	AT		
Zeile (3)				

☒ Das Anmeldeamt wird ersucht, eine beglaubigte Abschrift der oben in der (den) Zeile(n) 1 und 2 bezeichneten früheren Anmeldung(en) zu erstellen und dem internationalen Büro zu übermitteln (nur falls die frühere Anmeldung(en) bei dem Amt eingereicht worden ist(sind), das für die Zwecke dieser internationalen Anmeldung Anmeldeamt ist)

* Falls es sich bei der früheren Anmeldung um eine ARIPO-Anmeldung handelt, so muß in dem Zusatzfeld mindestens ein Staat angegeben werden, der Mitgliedstaat der Pariser Verbandsübereinkunft zum Schutz des gewerblichen Eigentums ist und für den die frühere Anmeldung eingereicht wurde.

Feld Nr. VII INTERNATIONALE RECHERCHENBEHÖRDE

Wahl der internationalen Recherchenbehörde (ISA)
(falls zwei oder mehr als zwei internationale Recherchenbehörden für die Ausführung der internationalen Recherche zuständig sind, geben Sie die von Ihnen gewählte Behörde an; der Zweibuchstaben-Code kann benutzt werden):

Antrag auf Nutzung der Ergebnisse einer früheren Recherche; Bezugnahme auf diese frühere Recherche (falls eine frühere Recherche bei der internationalen Recherchenbehörde beantragt oder von ihr durchgeführt worden ist):

Datum (Tag/Monat/Jahr) Aktenzeichen Staat (oder regionales Amt)

ISA /

Feld Nr. VIII KONTROLLISTE; EINREICHUNGSSPRACHE

Diese internationale Anmeldung enthält die folgende Anzahl von Blättern:

Antrag : 3
Beschreibung (ohne Sequenzprotokollteil) : 16
Ansprüche : 5
Zusammenfassung : 1
Zeichnungen : 6
Sequenzprotokollteil der Beschreibung :
Blattzahl insgesamt : 31

Dieser internationalen Anmeldung liegen die nachstehend angekreuzten Unterlagen bei:

- ☒ Blatt für die Gebührenberechnung
- ☐ Gesonderte unterzeichnete Vollmacht
- ☒ Kopie der allgemeinen Vollmacht; Aktenzeichen (falls vorhanden): 40704
- ☐ Begründung für das Fehlen einer Unterschrift
- ☒ Prioritätsbeleg(e), in Feld Nr. VI durch 1 und 2 folgende Zeilennummer gekennzeichnet:
- ☐ Übersetzung der internationalen Anmeldung in die folgende Sprache:
- ☐ Gesonderte Angaben zu hinterlegten Mikroorganismen oder anderem biologischen Material
- ☐ Protokoll der Nucleotid- und/oder Aminosäuresequenzen in computerlesbarer Form
- ☐ Sonstige (einzeln auflühren):

Abbildung der Zeichnungen, die mit der Zusammenfassung veröffentlicht werden soll (Nr.): 2

Sprache, in der die internationale Anmeldung eingereicht wird: Deutsch

Feld Nr. IX UNTERSCHRIFT DES ANMELDERS ODER DES ANWALTS

Der Name jeder unterzeichnenden Person ist neben der Unterschrift zu wiederholen, und es ist anzugeben, sofern sich dies nicht eindeutig aus dem Antrag ergibt, in welcher Eigenschaft die Person unterzeichnet.

SCHRÖDL Manfred Dipl. Ing. Dr.
vertreten durch

KRAUSE Peter
(VM Nr. 40709)

Vom Anmeldeamt auszufüllen	
1. Datum des tatsächlichen Eingangs dieser internationalen Anmeldung:	2. Zeichnungen <input type="checkbox"/> eingegangen: <input type="checkbox"/> nicht eingegangen:
3. Geändertes Eingangsdatum aufgrund nachträglich, jedoch fristgerecht eingegangener Unterlagen oder Zeichnungen zur Vervollständigung dieser internationalen Anmeldung:	
4. Datum des fristgerechten Eingangs der angeforderten Richtigstellungen nach Artikel 11(2) PCT:	
5. Internationale Recherchenbehörde (falls zwei oder mehr zuständig sind): ISA /	6. <input type="checkbox"/> Übermittlung des Recherchenexemplars bis zur Zahlung der Recherchegebühr aufgeschoben

Vom Internationalen Büro auszufüllen

Datum des Eingangs des Aktenexemplars beim Internationalen Büro:

PATENT COOPERATION TREATY

PCT

COMMUNICATION IN CASES FOR WHICH NO OTHER FORM IS APPLICABLE

From the INTERNATIONAL BUREAU

To:

KRAUSE, Peter
Sagerbachgasse 7
A-2500 Baden
AUTRICHE

Date of mailing (day/month/year) 21 November 2000 (21.11.00)	
Applicant's or agent's file reference pctS3	REPLY DUE see paragraph 1 below
International application No. PCT/AT00/00167	International filing date (day/month/year) 21 June 2000 (21.06.00)
Applicant SCHRODL, Manfred	

1. ☐ REPLY DUE within _____ months/days from the above date of mailing
- ☐ NO REPLY DUE, however, see below
- ☒ IMPORTANT COMMUNICATION
- ☐ INFORMATION ONLY

2. COMMUNICATION:

Following a communication from the receiving Office (RO/AT), the International Bureau confirms that the International Filing Date of the present international application has been corrected to read:

21 June 2000 (21.06.00)

instead of 21 July 2000 (21.07.00)

A copy of this communication is being sent to the receiving Office, the International Searching Authority and to the designated Offices that have already been informed of their designation.

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland	Authorized officer Ellen Moyse
Facsimile No. (41-22) 740.14.35	Telephone No. (41-22) 338.83.38

PATENT COOPERATION TREATY

PCT

NOTIFICATION CONCERNING
SUBMISSION OR TRANSMITTAL
OF PRIORITY DOCUMENT

(PCT Administrative Instructions, Section 411)

From the INTERNATIONAL BUREAU

To:

KRAUSE, Peter
Sagerbachgasse 7
A-2500 Baden
AUTRICHE

Date of mailing (day/month/year) 21 November 2000 (21.11.00)	IMPORTANT NOTIFICATION
Applicant's or agent's file reference pctS3	
International application No. PCT/AT00/00167	International filing date (day/month/year) 21 June 2000 (21.06.00)
International publication date (day/month/year) Not yet published	Priority date (day/month/year) 21 June 1999 (21.06.99)
Applicant SCHRÖDL, Manfred	

- The applicant is hereby notified of the date of receipt (except where the letters "NR" appear in the right-hand column) by the International Bureau of the priority document(s) relating to the earlier application(s) indicated below. Unless otherwise indicated by an asterisk appearing next to a date of receipt, or by the letters "NR", in the right-hand column, the priority document concerned was submitted or transmitted to the International Bureau in compliance with Rule 17.1(a) or (b).
- This updates and replaces any previously issued notification concerning submission or transmittal of priority documents.
- An asterisk (*) appearing next to a date of receipt, in the right-hand column, denotes a priority document submitted or transmitted to the International Bureau but not in compliance with Rule 17.1(a) or (b). In such a case, the attention of the applicant is directed to Rule 17.1(c) which provides that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity, upon entry into the national phase, to furnish the priority document within a time limit which is reasonable under the circumstances.
- The letters "NR" appearing in the right-hand column denote a priority document which was not received by the International Bureau or which the applicant did not request the receiving Office to prepare and transmit to the International Bureau, as provided by Rule 17.1(a) or (b), respectively. In such a case, the attention of the applicant is directed to Rule 17.1(c) which provides that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity, upon entry into the national phase, to furnish the priority document within a time limit which is reasonable under the circumstances.

<u>Priority date</u>	<u>Priority application No.</u>	<u>Country or regional Office or PCT receiving Office</u>	<u>Date of receipt of priority document</u>
21 June 1999 (21.06.99)	A 1081/99	AT	09 Augu 2000 (09.08.00)
15 Dec 1999 (15.12.99)	A 2115/99	AT	09 Augu 2000 (09.08.00)

The International Bureau of WIPO
34, chemin des Colombettes
1211 Geneva 20, Switzerland

Authorized officer:

Ellen Moyse

Facsimile No. (41-22) 740.14.35

Telephone No. (41-22) 338.83.38

PATENT COOPERATION TREATY

PCT

NOTICE INFORMING THE APPLICANT OF THE COMMUNICATION OF THE INTERNATIONAL APPLICATION TO THE DESIGNATED OFFICES

(PCT Rule 47.1(c), first sentence)

From the INTERNATIONAL BUREAU

To:
KRAUSE, Peter
Sagerbachgasse 7
A-2500 Baden
AUTRICHE

Date of mailing (day/month/year) 04 January 2001 (04.01.01)		IMPORTANT NOTICE	
Applicant's or agent's file reference pctS3			
International application No. PCT/AT00/00167	International filing date (day/month/year) 21 June 2000 (21.06.00)	Priority date (day/month/year) 21 June 1999 (21.06.99)	
Applicant SCHRÖDL, Manfred			

1. Notice is hereby given that the International Bureau has communicated, as provided in Article 20, the international application to the following designated Offices on the date indicated above as the date of mailing of this Notice:
AG,AU,BZ,DZ,KP,KR,MZ,US

In accordance with Rule 47.1(c), third sentence, those Offices will accept the present Notice as conclusive evidence that the communication of the international application has duly taken place on the date of mailing indicated above and no copy of the international application is required to be furnished by the applicant to the designated Office(s).

2. The following designated Offices have waived the requirement for such a communication at this time:
AE,AL,AM,AP,AZ,BA,BB,BG,BR,BY,CA,CH,CN,CR,CU,CZ,DE,DK,DM,EA,EE,EP,ES,FI,GB,GD,GE,
GH,GM,HR,HU,ID,IL,IN,IS,JP,KE,KG,KZ,LC,LK,LR,LS,LT,LU,LV,MA,MD,MG,MK,MN,MW,MX,NO,
NZ,OA,PL,PT,RO,RU,SD,SE,SG,SI,SK,SL,TJ,TM,TR,TT,TZ,UA,UG,UZ,VN,YU,ZA,ZW
The communication will be made to those Offices only upon their request. Furthermore, those Offices do not require the applicant to furnish a copy of the international application (Rule 49.1(a-bis)).
3. Enclosed with this Notice is a copy of the international application as published by the International Bureau on
04 January 2001 (04.01.01) under No. WO 01/01550

REMINDER REGARDING CHAPTER II (Article 31(2)(a) and Rule 54.2)

If the applicant wishes to postpone entry into the national phase until 30 months (or later in some Offices) from the priority date, a demand for international preliminary examination must be filed with the competent International Preliminary Examining Authority before the expiration of 19 months from the priority date.

It is the applicant's sole responsibility to monitor the 19-month time limit.
Note that only an applicant who is a national or resident of a PCT Contracting State which is bound by Chapter II has the right to file a demand for international preliminary examination.

REMINDER REGARDING ENTRY INTO THE NATIONAL PHASE (Article 22 or 39(1))

If the applicant wishes to proceed with the international application in the national phase, he must, within 20 months or 30 months, or later in some Offices, perform the acts referred to therein before each designated or elected Office.

For further important information on the time limits and acts to be performed for entering the national phase, see the Annex to Form PCT/IB/301 (Notification of Receipt of Record Copy) and Volume II of the PCT Applicant's Guide.

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland	Authorized officer J. Zahra
Facsimile No. (41-22) 740.14.35	Telephone No. (41-22) 338.83.38
Form PCT/IB/308 (July 1996)	

3743782

Continuation of Form PCT/IB/308

**NOTICE INFORMING THE APPLICANT OF THE COMMUNICATION OF
THE INTERNATIONAL APPLICATION TO THE DESIGNATED OFFICES**

Date of mailing (day/month/year) 04 January 2001 (04.01.01)	IMPORTANT NOTICE
Applicant's or agent's file reference pctS3	International application No. PCT/AT00/00167

The applicant is hereby notified that, at the time of establishment of this Notice, the time limit under Rule 46.1 for making amendments under Article 19 has not yet expired and the International Bureau had received neither such amendments nor a declaration that the applicant does not wish to make amendments.

P21760

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant : M. SCHROEDL

Appl No. : Not Yet Assigned

PCT Branch

I.A. Filed : June 21, 2000

PCT/ AT00/00167

For : ELECTRIC MOTOR

CLAIM OF PRIORITY

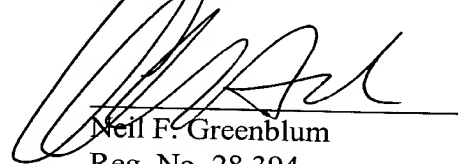
Commissioner of Patents and Trademarks

Washington, D.C. 20231

Sir:

Applicant hereby claims the right of priority granted pursuant to 35 U.S.C. 119 based upon Austrian Application No. A1081/99 filed 21 June 1999 and Application No. A2115/99 filed December 15, 1999. The International Bureau already should have sent certified copies of the Austrian applications to the United States designated office. If the certified copies have not arrived, please contact the undersigned.

Respectfully submitted,
M. SCHROEDL



Neil F. Greenblum
Reg. No. 28,394

December 20, 2001
GREENBLUM & BERNSTEIN, P.L.C.
1941 Roland Clarke Place
Reston, VA 20191
(703) 716-1191

Reg. No. 33,094

AUSTRIAN PATENT OFFICE

Ref.: A 1081/99-1,2

To Dr. Dipl.-Ing. Manfred Schrödl

Response dated June 25, 1999

1. Preliminary Report

The preliminary examination conducted according to § 99 of the Patent Act has produced the following result:

The subject matter of the present application is prejudiced by the following material made available to the general public before the priority date of the application:

- (D1) DE 28 14 884 A1 (GAP GESELLSCHAFT, October 11, 1979 (11.10.79), 16 pages)
Describes above all in claims 1, 2 and 6 through 8, as well as last para of page 5, middle of page 8 and middle para. of page 11, an electric motor such as that described in claims 1, 2 and 5 through 12 of the present application, anticipating it completely.
- (D2) WO 95/34117 A1 (PRECISE POWER CORPORATION, December 14, 1995 (14.12.95), 65 pages)
Describes above all in claims 1, 4, 6, 8 through 12, and in line 11 on page 13, line 17 on page 14 through line 10 on page 15, lines 6 through 20 on page 17, lines 10 through 23 on page 19, line 23 on page 19 through line 5 on page 20, lines 1 through 7 on page 23 and line 14 on page 30 through line 6 on page 31, an electric motor such as that described in claims 1 through 3 and 5 through 8 and 10, 11 and 13 of the present application, anticipating it completely.
- (D3) EP 0 320 415 A1 (RIVKINE, June 14 1989 (14.06.89), 9 pages)
Describes above all in claims 1 through 5, and in lines 11 through 47 of column 5, an electric motor such as that described in claims 1, 2, 5 and 13 of the present application, thus anticipating it completely again.
- (D4) EP 0 791 495 A2 (TOYOTA JIDOSHA, August 27, 1997 (27.08.97) 38 pages)
Describes above all in claims 1 through 3 and 13, 14 and in line 16 of column 4 through line 33 of column 7, and in line 16 of column 13 through line 36 of column 20, an electric motor such as that described in claims 1 through 3 of the present application, if the change of the battery voltage in (D4) is considered as "freely selectable voltage level" according to the present application. (D4) thus completely anticipates claims 1 through 3

P21760.TR2

of the present application again.

As a result, it is not discernible from the subject matter of the application what could constitute a patentable invention as defined by the provisions of §§ 1 and 3 of the Patent Act.

In connection with the problem and the solution that is to be accomplished with the subject matter of the present invention, no patentable difference is discernible in the features of claim 4—the only claim of this application not affected.

A patent cannot therefore be issued on the basis of the current documents.

[Instructions for response]

PATENT COOPERATION TREATY

PCT

NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

Commissioner
 US Department of Commerce
 United States Patent and Trademark
 Office, PCT
 2011 South Clark Place Room
 CP2/5C24
 Arlington, VA 22202
 ETATS-UNIS D'AMERIQUE
 in its capacity as elected Office

Date of mailing (day/month/year) 12 March 2001 (12.03.01)	Applicant's or agent's file reference pctS3
International application No. PCT/AT00/00167	
International filing date (day/month/year) 21 June 2000 (21.06.00)	Priority date (day/month/year) 21 June 1999 (21.06.99)
Applicant SCHRÖDL, Manfred	

1. The designated Office is hereby notified of its election made:

☒ in the demand filed with the International Preliminary Examining Authority on:
 15 January 2001 (15.01.01)

☐ in a notice effecting later election filed with the International Bureau on:

2. The election ☒ was

☐ was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO
 34, chemin des Colombettes
 1211 Geneva 20, Switzerland

Facsimile No.: (41-22) 740.14.35

Authorized officer

Henrik Nyberg

Telephone No.: (41-22) 338.83.38

VERTRAG ÜBER DIE INTERNATIONALE ZUSAMMENARBEIT AUF DEM GEBIET DES PATENTWESENS

Abseider: MIT DER INTERNATIONALEN VORLÄUFIGEN
PRÜFUNG BEAUFTRAGTE BEHÖRDE

An:

KRAUSE, Peter
Sagerbachgasse 7
A-2500 Baden
AUTRICHE

EINGANG

15. Okt. 2001

FRIST:

PCT

MITTEILUNG ÜBER DIE ÜBERSENDUNG
DES INTERNATIONALEN VORLÄUFIGEN
PRÜFUNGSBERICHTS

(Regel 71.1 PCT)

Absendedatum
(Tag/Monat/Jahr)

11.10.2001

Aktenzeichen des Anmelders oder Anwalts

pctS3

WICHTIGE MITTEILUNG

Internationales Aktenzeichen
PCT/AT00/00167

Internationales Anmeldedatum (Tag/Monat/Jahr)
21/06/2000

Prioritätsdatum (Tag/Monat/Jahr)
21/06/1999

Anmelder

SCHRÖDL MANFRED

1. Dem Anmelder wird mitgeteilt, daß ihm die mit der internationalen vorläufigen Prüfung beauftragte Behörde hiermit den zu der internationalen Anmeldung erstellten internationalen vorläufigen Prüfungsbericht, gegebenenfalls mit den dazugehörigen Anlagen, übermittelt.
2. Eine Kopie des Berichts wird - gegebenenfalls mit den dazugehörigen Anlagen - dem Internationalen Büro zur Weiterleitung an alle ausgewählten Ämter übermittelt.
3. Auf Wunsch eines ausgewählten Amtes wird das Internationale Büro eine Übersetzung des Berichts (jedoch nicht der Anlagen) ins Englische anfertigen und diesem Amt übermitteln.

4. ERINNERUNG

Zum Eintritt in die nationale Phase hat der Anmelder vor jedem ausgewählten Amt innerhalb von 30 Monaten ab dem Prioritätsdatum (oder in manchen Ämtern noch später) bestimmte Handlungen (Einreichung von Übersetzungen und Entrichtung nationaler Gebühren) vorzunehmen (Artikel 39 (1)) (siehe auch die durch das Internationale Büro im Formblatt PCT/IB/301 übermittelte Information).

Ist einem ausgewählten Amt eine Übersetzung der internationalen Anmeldung zu übermitteln, so muß diese Übersetzung auch Übersetzungen aller Anlagen zum internationalen vorläufigen Prüfungsbericht enthalten. Es ist Aufgabe des Anmelders, solche Übersetzungen anzufertigen und den betroffenen ausgewählten Ämtern direkt zuzuleiten.

Weitere Einzelheiten zu den maßgebenden Fristen und Erfordernissen der ausgewählten Ämter sind Band II des PCT-Leitfadens für Anmelder zu entnehmen.

Name und Postanschrift der mit der internationalen Prüfung beauftragten Behörde



Europäisches Patentamt - P.B. 5818 Patentlaan 2
NL-2280 HV Rijswijk - Pays Bas
Tel. +31 70 340 - 2040 Tx: 31 651 epo nl
Fax: +31 70 340 - 3016

Bevollmächtigter Bediensteter

Cardenas, C

Tel. +31 70 340-3370



VERTRAG ÜBER DIE INTERNATIONALE ZUSAMMENARBEIT AUF DEM GEBIET DES PATENTWESENS

PCT

INTERNATIONALER VORLÄUFIGER PRÜFUNGSBERICHT

(Artikel 36 und Regel 70 PCT)

Aktenzeichen des Anmelders oder Anwalts pctS3	WEITERES VORGEHEN siehe Mitteilung über die Übersendung des internationalen vorläufigen Prüfungsberichts (Formblatt PCT/IPEA/416)	
Internationales Aktenzeichen PCT/AT00/00167	Internationales Anmeldedatum (Tag/Monat/Jahr) 21/06/2000	Prioritätsdatum (Tag/Monat/Tag) 21/06/1999
Internationale Patentklassifikation (IPK) oder nationale Klassifikation und IPK H02K16/00		
Anmelder SCHRÖDL MANFRED		

1. Dieser internationale vorläufige Prüfungsbericht wurde von der mit der internationalen vorläufigen Prüfung beauftragten Behörde erstellt und wird dem Anmelder gemäß Artikel 36 übermittelt.



2. Dieser BERICHT umfaßt insgesamt 5 Blätter einschließlich dieses Deckblatts.

☒ Außerdem liegen dem Bericht ANLAGEN bei; dabei handelt es sich um Blätter mit Beschreibungen, Ansprüchen und/oder Zeichnungen, die geändert wurden und diesem Bericht zugrunde liegen, und/oder Blätter mit vor dieser Behörde vorgenommenen Berichtigungen (siehe Regel 70.16 und Abschnitt 607 der Verwaltungsrichtlinien zum PCT).

Diese Anlagen umfassen insgesamt 5 Blätter.

3. Dieser Bericht enthält Angaben zu folgenden Punkten:

- I ☒ Grundlage des Berichts
- II ☐ Priorität
- III ☐ Keine Erstellung eines Gutachtens über Neuheit, erfinderische Tätigkeit und gewerbliche Anwendbarkeit
- IV ☐ Mangelnde Einheitlichkeit der Erfindung
- V ☒ Begründete Feststellung nach Artikel 35(2) hinsichtlich der Neuheit, der erfinderischen Tätigkeit und der gewerblichen Anwendbarkeit; Unterlagen und Erklärungen zur Stützung dieser Feststellung
- VI ☐ Bestimmte angeführte Unterlagen
- VII ☒ Bestimmte Mängel der internationalen Anmeldung
- VIII ☐ Bestimmte Bemerkungen zur internationalen Anmeldung

Datum der Einreichung des Antrags 15/01/2001	Datum der Fertigstellung dieses Berichts 11.10.2001
Name und Postanschrift der mit der internationalen vorläufigen Prüfung beauftragten Behörde:  Europäisches Patentamt - P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk - Pays Bas Tel. +31 70 340 - 2040 Tx: 31 651 epo nl Fax: +31 70 340 - 3016	Bevollmächtigter Bediensteter Zoukas, E Tel. Nr. +31 70 340 3463 

INTERNATIONALER VORLÄUFIGER PRÜFUNGSBERICHT

Internationales Aktenzeichen PCT/AT00/00167

I. Grundlage des Berichts

1. Hinsichtlich der **Bestandteile** der internationalen Anmeldung (*Ersatzblätter, die dem Anmeldeamt auf eine Aufforderung nach Artikel 14 hin vorgelegt wurden, gelten im Rahmen dieses Berichts als "ursprünglich eingereicht" und sind ihm nicht beigefügt, weil sie keine Änderungen enthalten (Regeln 70.16 und 70.17)*):
Beschreibung, Seiten:

1-16 ursprüngliche Fassung

Patentansprüche, Nr.:

1-28 eingegangen am 03/08/2001 mit Schreiben vom 01/08/2001

Zeichnungen, Blätter:

1/6-6/6 ursprüngliche Fassung

2. Hinsichtlich der **Sprache**: Alle vorstehend genannten Bestandteile standen der Behörde in der Sprache, in der die internationale Anmeldung eingereicht worden ist, zur Verfügung oder wurden in dieser eingereicht, sofern unter diesem Punkt nichts anderes angegeben ist.

Die Bestandteile standen der Behörde in der Sprache: zur Verfügung bzw. wurden in dieser Sprache eingereicht; dabei handelt es sich um

- ☐ die Sprache der Übersetzung, die für die Zwecke der internationalen Recherche eingereicht worden ist (nach Regel 23.1(b)).
- ☐ die Veröffentlichungssprache der internationalen Anmeldung (nach Regel 48.3(b)).
- ☐ die Sprache der Übersetzung, die für die Zwecke der internationalen vorläufigen Prüfung eingereicht worden ist (nach Regel 55.2 und/oder 55.3).

3. Hinsichtlich der in der internationalen Anmeldung offenbarten **Nucleotid- und/oder Aminosäuresequenz** ist die internationale vorläufige Prüfung auf der Grundlage des Sequenzprotokolls durchgeführt worden, das:

- ☐ in der internationalen Anmeldung in schriftlicher Form enthalten ist.
- ☐ zusammen mit der internationalen Anmeldung in computerlesbarer Form eingereicht worden ist.
- ☐ bei der Behörde nachträglich in schriftlicher Form eingereicht worden ist.
- ☐ bei der Behörde nachträglich in computerlesbarer Form eingereicht worden ist.
- ☐ Die Erklärung, daß das nachträglich eingereichte schriftliche Sequenzprotokoll nicht über den Offenbarungsgehalt der internationalen Anmeldung im Anmeldezeitpunkt hinausgeht, wurde vorgelegt.
- ☐ Die Erklärung, daß die in computerlesbarer Form erfassten Informationen dem schriftlichen Sequenzprotokoll entsprechen, wurde vorgelegt.

4. Aufgrund der Änderungen sind folgende Unterlagen fortgefallen:

INTERNATIONALER VORLÄUFIGER PRÜFUNGSBERICHT

Internationales Aktenzeichen PCT/AT00/00167

- ☐ Beschreibung, Seiten:
☐ Ansprüche, Nr.:
☐ Zeichnungen, Blatt:

5. ☐ Dieser Bericht ist ohne Berücksichtigung (von einigen) der Änderungen erstellt worden, da diese aus den angegebenen Gründen nach Auffassung der Behörde über den Offenbarungsgehalt in der ursprünglich eingereichten Fassung hinausgehen (Regel 70.2(c)).

(Auf Ersatzblätter, die solche Änderungen enthalten, ist unter Punkt 1 hinzuweisen; sie sind diesem Bericht beizufügen).

6. Etwaige zusätzliche Bemerkungen:

V. Begründete Feststellung nach Artikel 35(2) hinsichtlich der Neuheit, der erfinderischen Tätigkeit und der gewerblichen Anwendbarkeit; Unterlagen und Erklärungen zur Stützung dieser Feststellung

1. Feststellung

Neuheit (N)	Ja: Ansprüche	1-28
	Nein: Ansprüche	
Erfinderische Tätigkeit (ET)	Ja: Ansprüche	1-28
	Nein: Ansprüche	
Gewerbliche Anwendbarkeit (GA)	Ja: Ansprüche	1-28
	Nein: Ansprüche	

2. Unterlagen und Erklärungen siehe Beiblatt

VII. Bestimmte Mängel der internationalen Anmeldung

Es wurde festgestellt, daß die internationale Anmeldung nach Form oder Inhalt folgende Mängel aufweist:
siehe Beiblatt

Zu Punkt V

Die Anmeldung betrifft eine elektrische Maschine, vorzugsweise in Drehstromausführung in der Kraftfahrzeugtechnik.

Als nächstkommender Stand der Technik wird D1 (EP-A-769403) angesehen.

Aus der D1 ist ein Hybrid-Antriebssystem bekannt (siehe Abbildungen 2,8), bei dem eine erste (16) und eine zweite (22) elektrische Maschine vorgesehen sind. Die erste elektrische Maschine ist über ihren Rotor mit einer rotierenden Welle einer Verbrennungskraftmaschine (12) mechanisch verbunden. Die zweite elektrische Maschine ist mit ihrem Rotor mit einem rotierenden Teil eines mechanischen Aggregates, in Form eines Getriebes (20), mechanisch gekuppelt.

Der Gegenstand des Anspruchs 1 unterscheidet sich von der Maschinen-anordnung des Dokuments D1 dadurch, dass:

die erste elektrische Maschine mit mindestens der zweiten elektrischen Maschine über leistungselektronische Elemente (12,13) zum **Austausch** elektrischer Energie auf **frei wählbarem** Spannungsniveau **elektrisch verbunden** ist.

Der Gegenstand des Anspruchs 1 ist somit neu.

Die unterscheidenden Merkmale ermöglichen einen eigenen, autarken, internen elektrischen Kreis, der unabhängig vom Spannungsniveau des Bordnetzes ist.

Die mit vorliegender Anmeldung zu lösende **Aufgabe** kann somit in der Angabe einer elektrischen Maschinen-anordnung gesehen werden, bei der der interne elektrische Kreis und damit das Spannungsniveau der ersten Maschine unabhängig von dem Spannungsniveau eines externen, elektrischen Kreises, des sogenannten Bordnetzes (14 auf Abbildung 2) ist.

Die im neuen Anspruch 1 (der sich aus dem ursprünglichen Anspruch 1 und dem auf Seite 11, Zeilen 4,5 dargestellten Merkmal zusammensetzt) vorgeschlagene **Lösung** ist neu und erfinderisch weil sich im Stand der Technik insgesamt keine Lehre findet, die den mit dem technischen Problem befassten Fachmann veranlassen würde, den nächstliegenden Stand der Technik (in dem beide Maschinen an das Batterienspannungsniveau gebunden sind) unter Berücksichtigung dieser Lehre zu ändern oder anzupassen und somit zu etwas zu gelangen, was unter den Patentanspruch fällt (d.h Maschinen die **aneinander elektrisch gekoppelt sind**), und das zu erreichen, was mit der Erfindung erreicht wird (**d.h Austausch** elektrischer Energie auf **frei wählbarem** Spannungsniveau).

In EP-A-0725474 und EP-A0800951 sind auch beide Maschinen an das Batterienspannungsniveau gebunden.

Der Gegenstand des unabhängigen Anspruchs 1 erfüllt somit die Erfordernisse der Artikel 33(2) , 33(3) PCT .

Die Ansprüche 2-28 sind abhängige Ansprüche, die im Zusammenhang mit dem Anspruch 1 die Erfordernisse der Artikel 33(2), 33(3) PCT erfüllen.

Die gewerbliche Anwendbarkeit der Gegenstände der Ansprüche 1-28 steht außer Zweifel. Damit erfüllen die Ansprüche 1-28 auch die Erfordernisse des Art. 33 (4) PCT.

Zu Punkt VII

1. Im Widerspruch zu den Erfordernissen der Regel 5.1 a) ii) PCT werden in der Beschreibung weder der in dem Dokument D1 offenbarte einschlägige Stand der Technik noch dieses Dokument angegeben.

2. a) Die Beschreibung steht nicht, wie in Regel 5.1 a) iii) PCT vorgeschrieben, in Einklang mit den Ansprüchen.

03. 08. 2001

Neue Patentansprüche:

(75)

1. Elektrische Maschine, vorzugsweise in Drehstromausführung, wobei eine
erste elektrische Maschine (10) vorgesehen ist, die über ihren Rotor (5) mit
5 einer rotierenden Welle einer Maschine, insbesondere einer
Verbrennungskraftmaschine, mechanisch verbunden ist und mindestens
eine zweite elektrische Maschine (11) vorgesehen ist, die mit ihrem Rotor
(6) mit einem rotierenden Teil eines mechanischen Aggregates mechanisch
gekuppelt ist, dadurch gekennzeichnet, daß die erste elektrische Maschine
10 (10) mit mindestens der zweiten elektrischen Maschine (11) über
leistungselektronische Elemente zum Austausch elektrischer Energie auf frei
wählbarem Spannungsniveau elektrisch verbunden ist.
2. Elektrische Maschine nach Anspruch 1, dadurch gekennzeichnet, daß die
15 zweite elektrische Maschine (11) mit ihrem Rotor (6) mit dem rotierenden
Teil einer Strömungsmaschine verbunden ist.
3. Elektrische Maschine nach Anspruch 1 oder 2, dadurch gekennzeichnet,
daß die erste elektrische Maschine (10) mit ihrem Rotor (5) mit einer
20 Kurbelwelle oder einer mit der Kurbelwelle in mechanischer Verbindung
stehenden Welle einer Verbrennungskraftmaschine mechanisch verbunden
ist.
4. Elektrische Maschine nach Anspruch 1 oder 2, dadurch gekennzeichnet,
25 daß die erste elektrische Maschine (10) mit der Verbrennungskraftmaschine
über ein Getriebe mechanisch verbunden ist.
5. Elektrische Maschine nach Anspruch 1 oder 2, dadurch gekennzeichnet,
daß die erste elektrische Maschine (10) ein Teil der
30 Verbrennungskraftmaschine ist, beispielsweise daß der Rotor (5) der ersten
elektrischen Maschine (10) in die Schwungscheibe der
Verbrennungskraftmaschine integriert ist.

6. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 5, dadurch gekennzeichnet, daß die erste elektrische Maschine (10) mit mindestens einem externen elektrischen Kreis, vorzugsweise einem Bordnetz (14), verbunden ist.

5

7. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 6, dadurch gekennzeichnet, daß die erste (10) und die zweite elektrische Maschine (11) in einem Gehäuse (9) angeordnet sind.

10 8. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 7, dadurch gekennzeichnet, daß die erste (10) und/oder die zweite elektrische Maschine (11) als Asynchron-, Synchron- oder Reluktanzmaschine ausgeführt ist bzw. sind.

15 9. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 8, dadurch gekennzeichnet, daß die erste (10) und die zweite elektrische Maschine (11) Rotoren (5, 6) mit gleicher Rotationsachse aufweisen.

20 10. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 9, dadurch gekennzeichnet, daß eine der beiden Maschinen (10, 11) als Innenläufer und die andere Maschine als Außenläufer ausgeführt sind.

25 11. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 10, dadurch gekennzeichnet, daß die beiden elektrischen Maschinen (10, 11) ein gemeinsames Statorblechpaket aufweisen.

30 12. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 11, dadurch gekennzeichnet, daß die Komponenten für den elektrischen Energieaustausch zwischen den elektrischen Maschinen (10, 11) und/oder einem externen elektrischen Kreis (14) in einem Gehäuse (9) mindestens einer elektrischen Maschine (10, 11) angeordnet sind.

13. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 12, dadurch gekennzeichnet, daß das Gehäuse (9) mindestens einer elektrischen Maschine (10, 11) eine Flüssigkeitskühlung aufweist.
- 5 14. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 13, dadurch gekennzeichnet, daß vom elektrischen Kreis, der die beiden elektrischen Maschinen verbindet, ein Netzanschluß mit Gleich-, Wechsel- oder Drehspannung ableitbar ist.
- 10 15. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 14, dadurch gekennzeichnet, daß der Stator (1, 4) mindestens einer elektrischen Maschine (10, 11) mindestens zwei, vorzugsweise in der Maschine (10, 11) galvanisch getrennte, Wicklungssysteme (22, 23) aufweist, die mit dem Hauptfluß der Maschine (10, 11) magnetisch
- 15 gekoppelt sind.
16. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 15, dadurch gekennzeichnet, daß die mindestens zwei Wicklungssysteme (22, 23) über getrennte leistungselektronische Schaltungen (24, 25) mit
- 20 jeweiligen, vorzugsweise galvanisch getrennten, Stromkreisen verbunden sind.
17. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 16, dadurch gekennzeichnet, daß mindestens ein Wicklungssystem (22, 23)
- 25 über eine Gleichrichterbrücke mit einem Gleichspannungs- oder batteriegestützten Netz, vorzugsweise einem Bordnetz (26), zum Energieaustausch in einer Richtung verbunden ist.
18. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 17,
- 30 dadurch gekennzeichnet, daß mindestens ein Wicklungssystem (22, 23) über eine Transistorbrücke mit einem Gleichspannungs- oder

zwischen diesen Wicklungssystemen (22, 23) unabhängig von einer Rotordrehung nach dem Prinzip des Transformators gegeben ist.

5 24. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 23, dadurch gekennzeichnet, daß durch schwache magnetische Koppelung von den Wicklungssystemen (22, 23) eine geringe elektromagnetische Beeinflussung der Wicklungssysteme (22, 23) erfolgt.

10 25. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 24, dadurch gekennzeichnet, daß durch Steuerung der elektromagnetischen Größen, vorzugsweise der Ströme und Flussverkettungen, mindestens eines Wicklungssystems (22, 23) ein beliebig gestaltbarer elektromechanischer Energieaustausch zwischen den Wicklungssystemen (22, 23) und der Rotorwelle erreichbar ist.

15 26. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 25, dadurch gekennzeichnet, daß eine erste und die zweite elektrische Maschine (10, 11) in einem Gehäuse angeordnet sind.

20 27. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 26, dadurch gekennzeichnet, daß die erste und/oder die zweite elektrische Maschine (10, 11) als Asynchron-, Synchron- oder Reluktanzmaschine ausgeführt ist bzw. sind.

25 28. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 27, dadurch gekennzeichnet, daß die erste und die zweite elektrische Maschine (10, 11) Rotoren mit gleicher Rotationsachse aufweisen.

30

Schrödl Manfred
vertreten durch
Krause Peter
(VM Nr. 40709)





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ANMELDETAG: 1999 12 15 GESCHÄFTSZAHLE: A 2115/99 -1
IPC: H02K (IN ALLEN EINGABEN ANFÜHREN)
AN SCHRÖDL MANFRED DIPL. ING. DR.
IN *OB* A-7223 SIECORABEN
UNTERE HAUPTSTRASSE 9

FRIST: 21.11.00

Eingabe vom 20. Dezember 1999

1. Vorbescheid

Die gemäß § 99 des Patentgesetzes vorgenommene Vorprüfung hat das unten stehende Ergebnis geliefert:

Dem Gegenstand der vorliegenden Anmeldung steht folgendes, der Öffentlichkeit vor dem Prioritätstag der Anmeldung zugänglich gemachtes Material entgegen:
US 4 117 390 A (Iwata et al.) 26. September 1978, (29.09.78), siehe gesamtes Dokument, US 5 412 268 A (Amaud et al.) 2. Mai 1995, (02.05.95), siehe gesamtes Dokument.

Patents Abstracts of Japan Vol. 006 N° 211 (23. Oktober 1982 (23.10.82))
& JP 5711 6578 A (Nippon Koguyu Tetsudo) 20. Juli 1982, (20.07.82),
Patents Abstracts of Japan Vol. 97 N° 07 (31. Juli 1997 (31.07.97)),
& JP 9 074 701 A (Toyoto Jidosha), 18. März 1997, (18.03.97)

Die genannten Dokumente zeigen jeweils Motor-Generator Ausführungen mit zwei getrennt steuerbaren Wicklungssystemen im Stator mit Energieaustausch auf wählbarem Spannungsniveau, merkmalsmäßig dem vorliegenden Anspruch 1 entsprechend.

Auch lassen die vorliegenden Unteransprüche im Hinblick auf die Entgegenhaltungen patentbegründende Merkmale nicht erblicken. Für die vorliegende Anspruchsfassung kann somit eine Erlangung eines Patents keinesfalls in Aussicht gestellt werden.

Sie werden eingeladen, sich innerhalb einer Frist von zwei Monaten ab Zustellung dieses Vorbescheides hierüber zu äußern bzw. innerhalb dieser Frist die angeführten Mängel Ihrer Anmeldung zu beheben.

Wird innerhalb dieser Frist weder den erteilten Aufträgen entsprochen, noch eine Äußerung oder ein Antrag auf Verlängerung der Frist überreicht, so gilt die Anmeldung als zurückgenommen. Diese Rechtsfolge tritt außer Kraft, wenn binnen vier Monaten nach Ablauf der Frist den erteilten Aufträgen entsprochen (bzw. die Äußerung auf den Vorbescheid nachgeholt) und eine Gebühr im Ausmaß der Anmeldegebühr (700 S) [50,87 €] auf das Postscheckkonto Nr. 5.160.000 des Patentamtes eingezahlt wird. Der Antrag auf Verlängerung der Frist unterliegt einer Verfahrensgebühr in der Höhe von 170 S (12,35 €), die nicht in Stempelmarken entrichtet werden darf, sondern auf das Postscheckkonto Nr. 5.160.000 des Patentamtes eingezahlt werden muss.

Wird ein nummerierter Erlagschein des Patentamtes verwendet, kann die Zahlung der oben angeführten Gebühren durch Überreichung der Auftragsbestätigung entweder im Original oder in Kopie nachgewiesen werden, andernfalls ist der urschriftliche Einzahlungs- oder Überweisungsbeleg vorzulegen.

Der Antrag auf Verlängerung der Frist ist als Eingabe zu stempeln.

Österreichisches Patentamt

Technische Abteilung XV

Wien, am 14. Juli 2000

Dipl.-Ing. Schlechter

P21760.TR1

Office Action

International Ref.: PCT/AT00/00167

I. Basis for the Office Action

1. Concerning the components of the International Application:

Specification, pages:

1-16 Original version

Claims, no.:

1-27 Original version

Drawings, sheets:

1/6-6/6 Original version

V. Substantiated determination of novelty, inventive activity and usefulness according to Article 35(2); documents and explanations to support this determination

1. Determination

Novelty Claims

Inventive Activity Claims

Usefulness Claims

2. Documents and explanations:

See annex

VII. Defects in the International Application

It has been established that the international application has the following defects in form or content:

See annex

Section V

1. The combination of features contained in independent claim 1 is not known from or made obvious by the prior art. Claim 1 and claims 2-27, which are dependent on claim 1, therefore meet the requirements of the PCT with regard to novelty and inventive activity. However, the application does not meet the requirements of Article 6 of the PCT because claim 1 is not clear (see Section VII).

Section VII

1. Although claim 1 is formulated in the two-part form, the features that are defined in the first nine lines of the claim (i.e. the first electric motor (16), the internal-combustion engine (12) and the second electric motor (22)) are incorrectly listed in the characterizing portion. However, these features have already been disclosed in document D1: EP-A-769403 (see Figs. 1, 2, 8; column 8, lines 47-53, column 2, lines 18-22). (Rule 6.3 b) ii) PC)
2. In order to meet the requirements of Rule 5.1 a) ii) PC, the above-mentioned document D1 should be cited in the specification and the relevant prior art it contains should be briefly outlined.
3. If the claims are amended, the specification must also be adjusted to the amended claims (Rule 5.1 (a) (iii) PC).
4. In order to facilitate the examination of amended application documents in terms of Article 34 (2) b) PC, the applicant is requested to clearly indicate the amendments made, regardless of whether these amendments are additions, replacements or deletions, and to state which parts of the application as originally filed these amendments are based on (see also Rule 66.8 a) PC).

If necessary, this information can be provided in handwritten form on copies of the relevant parts of the original application.

SCHRIFTLICHER BESCHEID

Internationales Aktenzeichen PCT/AT00/00167

I. Grundlage des Bescheids

1. Hinsichtlich der **Bestandteile** der internationalen Anmeldung (*Ersatzblätter, die dem Anmeldeamt auf eine Aufforderung nach Artikel 14 hin vorgelegt wurden, gelten im Rahmen dieses Bescheids als "ursprünglich eingereicht"*):

Beschreibung, Seiten:

1-16 ursprüngliche Fassung

Patentansprüche, Nr.:

1-27 ursprüngliche Fassung

Zeichnungen, Blätter:

1/6-6/6 ursprüngliche Fassung

2. Hinsichtlich der **Sprache**: Alle vorstehend genannten Bestandteile standen der Behörde in der Sprache, in der die internationale Anmeldung eingereicht worden ist, zur Verfügung oder wurden in dieser eingereicht, sofern unter diesem Punkt nichts anderes angegeben ist.

Die Bestandteile standen der Behörde in der Sprache: zur Verfügung bzw. wurden in dieser Sprache eingereicht; dabei handelt es sich um

- ☐ die Sprache der Übersetzung, die für die Zwecke der internationalen Recherche eingereicht worden ist (nach Regel 23.1(b)).
- ☐ die Veröffentlichungssprache der internationalen Anmeldung (nach Regel 48.3(b)).
- ☐ die Sprache der Übersetzung, die für die Zwecke der internationalen vorläufigen Prüfung eingereicht worden ist (nach Regel 55.2 und/oder 55.3).

3. Hinsichtlich der in der internationalen Anmeldung offenbaren **Nucleotid- und/oder Aminosäuresequenz** ist die internationale vorläufige Prüfung auf der Grundlage des Sequenzprotokolls durchgeführt worden, das:

- ☐ in der internationalen Anmeldung in schriftlicher Form enthalten ist.
- ☐ zusammen mit der internationalen Anmeldung in computerlesbarer Form eingereicht worden ist.
- ☐ bei der Behörde nachträglich in schriftlicher Form eingereicht worden ist.
- ☐ bei der Behörde nachträglich in computerlesbarer Form eingereicht worden ist.
- ☐ Die Erklärung, daß das nachträglich eingereichte schriftliche Sequenzprotokoll nicht über den Offenbarungsgehalt der internationalen Anmeldung im Anmeldezeitpunkt hinausgeht, wurde vorgelegt.
- ☐ Die Erklärung, daß die in computerlesbarer Form erfassten Informationen dem schriftlichen Sequenzprotokoll entsprechen, wurde vorgelegt.

SCHRIFTLICHER BESCHEID

Internationales Aktenzeichen PCT/AT00/00167

4. Aufgrund der Änderungen sind folgende Unterlagen fortgefallen:

- ☐ Beschreibung, Seiten:
- ☐ Ansprüche, Nr.:
- ☐ Zeichnungen, Blatt:

5. ☐ Dieser Bericht ist ohne Berücksichtigung (von einigen) der Änderungen erstellt worden, da diese aus den angegebenen Gründen nach Auffassung der Behörde über den Offenbarungsgehalt in der ursprünglich eingereichten Fassung hinausgehen (Regel 70.2(c)).

(Auf Ersatzblätter, die solche Änderungen enthalten, ist unter Punkt 1 hinzuweisen; sie sind diesem Bericht beizufügen.)

6. Etwaige zusätzliche Bemerkungen:

V. Begründete Feststellung nach Regel 66.2(a)(ii) hinsichtlich der Neuheit, der erfinderischen Tätigkeit und der gewerblichen Anwendbarkeit; Unterlagen und Erklärungen zur Stützung dieser Feststellung

- | | |
|--------------------------------|-----------|
| 1. Feststellung | |
| Neuheit (N) | Ansprüche |
| Erfinderische Tätigkeit (IS) | Ansprüche |
| Gewerbliche Anwendbarkeit (IA) | Ansprüche |

2. Unterlagen und Erklärungen:
siehe Beiblatt

VII. Bestimmte Mängel der internationalen Anmeldung

Es wurde festgestellt, daß die internationale Anmeldung nach Form oder Inhalt folgende Mängel aufweist:
siehe Beiblatt

**SCHRIFTLICHER BESCHEID
BEIBLATT**

Internationales Aktenzeichen PCT/AT00/00167

Änderungen, unabhängig davon, ob es sich um Änderungen durch Hinzufügen, Ersetzen oder Streichen handelt, deutlich aufzuzeigen und anzugeben, auf welche Stellen in der ursprünglich eingereichten Anmeldung sich diese Änderungen stützen (siehe auch Regel 66.8 a) PC).

Gegebenenfalls können diese Angaben in handschriftlicher Form auf Kopien der betreffenden Teile der ursprünglichen Anmeldung erfolgen.

12/04 02 10.41. FMA 02204 000
SCHRIFTLICHER BESCHEID
BEIBLATT

Internationales Aktenzeichen PCT/AT00/00167

Zu Punkt V

1. Die im unabhängigen Anspruch 1 enthaltene **Merkmalskombination** ist aus dem vorliegenden Stand der Technik weder bekannt, noch wird sie durch ihn nahegelegt. Anspruch 1 und die Ansprüche 2-27 die vom Anspruch 1 abhängig sind, erfüllen damit die Erfordernisse des PCT in Bezug auf Neuheit und erfinderische Tätigkeit. Die Anmeldung erfüllt aber nicht die Erfordernisse des Artikels 6 PCT, weil der Anspruch 1 nicht klar ist. (siehe Punkt VII).

Zu Punkt VII

1. Der Anspruch 1 ist zwar in der zweiteiligen Form abgefasst; die Merkmale, die auf den ersten neun Zeilen des Anspruchs definiert sind (d.h. die erste elektrische Maschine (16), die Verbrennungskraftmaschine (12) und die zweite elektrische Maschine (22)), sind aber unrichtigerweise im kennzeichnenden Teil aufgeführt. Diese Merkmale sind aber bereits im Dokument D1: EP-A-769403 (siehe Abbildungen 1, 2, 8; Spalte 8, Zeilen 47-53, Spalte 2, Zeilen 18-22) offenbart worden. (Regel 6.3 b) ii) PC).

2. Damit die Erfordernisse der Regel 5.1 a) ii) PC erfüllt werden, sollte in der Beschreibung das oben erwähnte Dokument D1 angegeben werden und der darin enthaltene einschlägige Stand der Technik sollte kurz umrissen werden.

3. Bei einer Änderung der Ansprüche müsste die Beschreibung auch an die geänderten Ansprüche angepasst werden (Regel 5.1 (a) (iii) PC).

4. Um die Prüfung von geänderten Anmeldungsunterlagen im Hinblick auf Artikel 34(2) b) PC zu erleichtern, wird der Anmelder gebeten, die durchgeführten

P21760.TR3

AUSTRIAN PATENT OFFICE

Ref.: A 2115/99-1

To: Dr. Dipl.-Ing. Manfred Schrödl

Response dated December 20, 1999

1. Preliminary Report

The preliminary examination conducted according to § 99 of the Patent Act has produced the following result:

The subject matter of the present application is prejudiced by the following material made available to the general public before the priority date of the application:

US 4 117 390 A (Iwata et al.) September 26, 1978 (29.09.78), see entire document, US 5 412 268 A (Arnaud et al.) May 2, 1995, (02.05.95), see entire document.

Patents Abstracts of Japan Vol. 006 No. 211 (October 23, 1982 (23.10.82))

& JP 5711 6578 A (Nippon Koguyu Tetsudo) July 20, 1982 (20.07.82),

Patents Abstracts of Japan Vol. 97 No. 07 (July 31, 1997 (31.07.97)),

& JP 9 074 701 A (Toyoto Jidosha), March 18, 1997 (18.03.97)

The referenced documents each show motor-generator embodiments with two separately controllable winding systems in the stator with energy exchange at a selectable voltage level, corresponding to the present claim 1 in terms of features.

The present subordinate claims do not disclose patentable features in view of the references, either. Therefore a patent cannot be issued for the present version of the claims.

[Instructions for response]

13-Mär-01 09:27 TU-WIEN E372

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 UID-Nr. ATU38265407; DVR: 0078018

ANMELDETAG: 1999 06 21

IPC: H02K

AN SCHRÖDL MANFRED DIPL. ING. DR.

IN *OB* A-7223 SIEGGRABEN

UNTERE HAUPTSTRASSE 9

GESCHÄFTSZAHL: A 1081/99-1,2
 (IN ALLEN EINGABEN ANFÜHREN)

EINGANG

13. März 2001

FRIST

1. Vorbescheid

Eingabe vom 25. Juni 1999

Die gemäß § 99 des Patentgesetzes vorgenommene Vorprüfung hat das unten stehende Ergebnis geliefert:

Dem Gegenstand der vorliegenden Anmeldung steht folgendes, der Öffentlichkeit vor dem Prioritätstag der Anmeldung zugänglich gemachtes Material entgegen:

- (L1) ... DE 28 14 884 A1 (GAP GESELLSCHAFT, 11. Oktober 1979 (11.10.79), 16 Seiten)
 beschreibt vor allem mit den Ansprüchen 1, 2 und 6 bis 8, sowie mit Seite 5 letzter Absatz, Seite 8 Mitte und Seite 11, mittlerer Absatz, eine elektrische Maschine, so wie es die Ansprüche 1, 2 und 5 bis 12 der vorliegenden Anmeldung beschreiben und nehmen diese daher zur Gänze vorweg.
- (L2) ... WO 95/34117 A1 (PRECISE POWER CORPORATION, 14. Dezember 1995 (14.12.95), 65 Seiten)
 beschreibt vor allem mit den Ansprüchen 1, 4, 6, 8 bis 12, sowie mit Seite 13 Zeile 11, Seite 14 Zeile 17 bis Seite 15 Zeile 10, Seite 17 Zeile 6 bis 20, Seite 19 Zeile 10 bis 23, Seite 19 Zeile 23 bis Seite 20 Zeile 5, Seite 23 Zeile 1 bis 7 und Seite 30 Zeile 14 bis Seite 31 Zeile 6, eine elektrische Maschine, so wie es die Ansprüche 1 bis 3 und 5 bis 8 sowie 10, 11 und 13 der vorliegenden Anmeldung beschreiben und nehmen diese daher neuerlich zur Gänze vorweg.
- (L3) ... EP 0 320 415 A1 (RIVKINE, 14. Juni 1989 (14.06.89), 9 Seiten)
 beschreibt vor allem mit den Ansprüchen 1 bis 5, sowie in Spalte 5 Zeile 11 bis 47, eine elektrische Maschine, so wie es die Ansprüche 1, 2, 5 und 13 der vorliegenden Anmeldung beschreiben und nehmen diese daher ein weiteres Mal zur Gänze vorweg.
- (L4) ... EP 0 791 495 A2 (TOYOTA JIDOSHA, 27. August 1997 (27.08.97), 38 Seiten)
 beschreibt vor allem mit den Ansprüchen 1 bis 3 und 13, 14, sowie in Spalte 4 Zeile 15 bis Spalte 7 Zeile 33 und auch in Spalte 13 Zeile 16 bis Spalte 20 Zeile 36, eine elektrische Maschine, so wie es die Ansprüche 1 bis 3 der vorliegenden Anmeldung beschreiben, wenn die Änderung der Batteriespannung in (L4), entsprechend der vorliegenden Anmeldung als "frei wählbares Spannungsniveau" angesehen wird. (L4) nimmt dann die Ansprüche 1 bis 3 der vorliegenden Anmeldung neuerlich zur Gänze vorweg.

Auf Grund dieses Sachverhaltes ist beim Anmeldungsgegenstand nicht erkennbar, worin noch eine patentierbare Erfindung im Sinne der Bestimmungen der §§ 1 und 3 PatG gelegen sein könnte.

Frei 12.5.01
 gemäß OPA!
 Freigegeben 13.3.01

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S.02

Denn in den Merkmalen des einzigen nicht getroffenen Anspruchs 4 der vorliegenden Anmeldung kann, im Zusammenhang mit der Aufgabenstellung und der Lösung, die mit dem vorliegenden Anmeldegegenstand bewerkstelligt werden soll, kein patentierbarer Überschuss erkannt werden.

Mit der Erteilung eines Patentes kann daher auf Grund der derzeit gegebenen Aktenlage nicht gerechnet werden.

Bei Weiterverfolgung der Anmeldung trotz obiger Bedenken wird Ihnen freigestellt, sich innerhalb einer Frist von zwei Monaten ab Zustellung dieses Vorbescheides hierüber zu äußern.

Wird innerhalb dieser Frist weder eine Äußerung oder ein Antrag auf Verlängerung der Frist überreicht, so gilt die Anmeldung als zurückgenommen. Diese Rechtsfolge tritt außer Kraft, wenn binnen vier Monaten nach Ablauf der Frist die Äußerung auf den Vorbescheid nachgeholt und eine Gebühr im Ausmaß der Anmeldegebühr (700 S) (50,87 €) auf das Postscheckkonto Nr. 5.160.000 des Patentamtes eingezahlt wird. Der Antrag auf Verlängerung der Frist unterliegt einer Verfahrensgebühr in der Höhe von 170 S (12,35 €), die nicht in Stempelmarken entrichtet werden darf, sondern auf das Postscheckkonto Nr. 5.160.000 des Patentamtes eingezahlt werden muss.

Wird ein nummerierter Erlagschein des Patentamtes verwendet, kann die Zahlung der oben angeführten Gebühren durch Überreichung der Auftragsbestätigung entweder im Original oder in Kopie nachgewiesen werden, andernfalls ist der urschriftliche Einzahlungs- oder Überweisungsbeleg vorzulegen. Der Antrag auf Verlängerung der Frist ist als Eingabe zu stempeln.

Österreichisches Patentamt

Technische Abteilung I

Wien, am 16. Feber 2001

(Dipl. Ing. Hawel)

Dipl. Ing. Ludwig

Translation

PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

9/926792

Applicant's or agent's file reference pctS3	FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/AT00/00167	International filing date (day/month/year) 21 June 2000 (21.06.00)	Priority date (day/month/year) 21 June 1999 (21.06.99)
International Patent Classification (IPC) or national classification and IPC H02K 16/00		
Applicant SCHRÖDL, Manfred		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 5 sheets, including this cover sheet.

☒ This report is also accompanied by ANNEXES, i.e., sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 5 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☒ Certain defects in the international application
- VIII ☐ Certain observations on the international application

Date of submission of the demand 15 January 2001 (15.01.01)	Date of completion of this report 11 October 2001 (11.10.2001)
Name and mailing address of the IPEA/EP	Authorized officer
Facsimile No.	Telephone No.

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No.

PCT/AT00/00167

I. Basis of the report

1. With regard to the **elements** of the international application:*

- ☐ the international application as originally filed
- ☒ the description:
 pages 1-16, as originally filed
 pages _____, filed with the demand
 pages _____, filed with the letter of _____
- ☒ the claims:
 pages _____, as originally filed
 pages _____, as amended (together with any statement under Article 19
 pages _____, filed with the demand
 pages 1-28, filed with the letter of 03 August 2001 (03.08.2001)
- ☒ the drawings:
 pages 1/6-6/6, as originally filed
 pages _____, filed with the demand
 pages _____, filed with the letter of _____
- ☐ the sequence listing part of the description:
 pages _____, as originally filed
 pages _____, filed with the demand
 pages _____, filed with the letter of _____

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language _____ which is:

- ☐ the language of a translation furnished for the purposes of international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of the translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. ☐ The amendments have resulted in the cancellation of:

- ☐ the description, pages _____
- ☐ the claims, Nos. _____
- ☐ the drawings, sheets/fig _____

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).**

* Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rule 70.16 and 70.17).

** Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No.
PCT/AT 00/00167

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Claims	1-28	YES
	Claims		NO
Inventive step (IS)	Claims	1-28	YES
	Claims		NO
Industrial applicability (IA)	Claims	1-28	YES
	Claims		NO

2. Citations and explanations

The application concerns an electric motor, preferably of the three-phase current design in the automobile industry.

EP-A-0 769 403 (D1) is considered the closest prior art. D1 describes a hybrid propulsion system (see Figures 2 and 8) in which a first (16) and a second (22) electric motors are provided. The rotor of the first electric motor is mechanically coupled to a rotating shaft of an internal combustion engine (12). The rotor of the second electric motor is mechanically coupled to a rotating part of a mechanical assembly in the form of a transmission (20). The subject matter of Claim 1 differs from the motor arrangement of D1 in that the first electric motor is **electrically connected** to at least the second electric motor by power electronic elements (12, 13) for **exchanging** electric energy **on a freely selectable** voltage level.

The subject matter of Claim 1 is therefore novel.

The distinguishing features achieve an independent, autonomous internal electric circuit independent of the voltage level of the board network. The present invention can therefore be considered to address the **problem** of providing an electric motor arrangement in which the

internal electric circuit and hence the voltage level of the first motor is independent of the voltage level of an external electric circuit, the so-called board network (14 in Figure 2).

The **solution** proposed in the new Claim 1 (composed of the original Claim 1 and of the feature described on page 11, lines 4 and 5) is novel and inventive because there is no teaching in the totality of the prior art which would prompt a person skilled in the art dealing with this technical problem to modify or adapt the closest prior art (in which the two motors are connected to the battery voltage level), taking into account this teaching, and hence to arrive at a solution covered by the present claim (i.e. motors which **are electrically coupled to one another**) and to achieve the same results as the invention (i.e. **the exchange of electric energy on a freely selectable voltage level**).

In EP-A-0 725 474 and EP-A-0 800 951, the two motors are also connected to the battery voltage level.

The subject matter of independent Claim 1 therefore meets the requirements of PCT Article 33(2) and (3).

Claims 2-28 are dependent claims which, in conjunction with Claim 1, meet the requirements of PCT Article 33(2) and (3).

The subjects of Claims 1-28 are unquestionably industrially applicable. Claims 1-28 therefore also meet the requirements of PCT Article 33(4).

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No.

PCT/AT 00/00167

VII. Certain defects in the international application

The following defects in the form or contents of the international application have been noted:

1. Contrary to PCT Rule 5.1(a)(ii), the description does not cite document D1 or indicate the relevant prior art disclosed therein.
2. Contrary to PCT Rule 5.1(a)(iii), the description is not in line with the claims.

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VERTRAG ÜBER DIE INTERNATIONALE ZUSAMMENARBEIT AUF DEM GEBIET DES PATENTWESENS

PCT

REC'D 12 OCT 2001

INTERNATIONALER VORLÄUFIGER PRÜFUNGSBERICHT

(Artikel 36 und Regel 70 PCT)

Aktenzeichen des Anmelders oder Anwalts pctS3	WEITERES VORGEHEN siehe Mitteilung über die Übersendung des internationalen vorläufigen Prüfungsberichts (Formblatt PCT/IPEA/416)	
Internationales Aktenzeichen PCT/AT00/00167	Internationales Anmeldedatum (Tag/Monat/Jahr) 21/06/2000	Prioritätsdatum (Tag/Monat/Jahr) 21/06/1999
Internationale Patentklassifikation (IPK) oder nationale Klassifikation und IPK H02K16/00		
Anmelder SCHRÖDL MANFRED		



1. Dieser internationale vorläufige Prüfungsbericht wurde von der mit der internationalen vorläufigen Prüfung beauftragten Behörde erstellt und wird dem Anmelder gemäß Artikel 36 übermittelt.
2. Dieser BERICHT umfaßt insgesamt 5 Blätter einschließlich dieses Deckblatts.

☒ Außerdem liegen dem Bericht ANLAGEN bei; dabei handelt es sich um Blätter mit Beschreibungen, Ansprüchen und/oder Zeichnungen, die geändert wurden und diesem Bericht zugrunde liegen, und/oder Blätter mit vor dieser Behörde vorgenommenen Berichtigungen (siehe Regel 70.16 und Abschnitt 607 der Verwaltungsrichtlinien zum PCT).

Diese Anlagen umfassen insgesamt 5 Blätter.

3. Dieser Bericht enthält Angaben zu folgenden Punkten:

- I ☒ Grundlage des Berichts
- II ☐ Priorität
- III ☐ Keine Erstellung eines Gutachtens über Neuheit, erfinderische Tätigkeit und gewerbliche Anwendbarkeit
- IV ☐ Mangelnde Einheitlichkeit der Erfindung
- V ☒ Begründete Feststellung nach Artikel 35(2) hinsichtlich der Neuheit, der erfinderischen Tätigkeit und der gewerblichen Anwendbarkeit; Unterlagen und Erklärungen zur Stützung dieser Feststellung
- VI ☐ Bestimmte angeführte Unterlagen
- VII ☒ Bestimmte Mängel der internationalen Anmeldung
- VIII ☐ Bestimmte Bemerkungen zur internationalen Anmeldung

Datum der Einreichung des Antrags 15/01/2001	Datum der Fertigstellung dieses Berichts 11.10.2001
Name und Postanschrift der mit der internationalen vorläufigen Prüfung beauftragten Behörde:  Europäisches Patentamt - P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk - Pays Bas Tel. +31 70 340 - 2040 Tx: 31 651 epo nl	Bevollmächtigter Bediensteter Zoukas, E 

I. Grundlage des Berichts

1. Hinsichtlich der **Bestandteile** der internationalen Anmeldung (*Ersatzblätter, die dem Anmeldeamt auf eine Aufforderung nach Artikel 14 hin vorgelegt wurden, gelten im Rahmen dieses Berichts als "ursprünglich eingereicht" und sind ihm nicht beigelegt, weil sie keine Änderungen enthalten (Regeln 70.16 und 70.17)*):
Beschreibung, Seiten:

1-16 ursprüngliche Fassung

Patentansprüche, Nr.:

1-28 eingegangen am 03/08/2001 mit Schreiben vom 01/08/2001

Zeichnungen, Blätter:

1/6-6/6 ursprüngliche Fassung

2. Hinsichtlich der **Sprache**: Alle vorstehend genannten Bestandteile standen der Behörde in der Sprache, in der die internationale Anmeldung eingereicht worden ist, zur Verfügung oder wurden in dieser eingereicht, sofern unter diesem Punkt nichts anderes angegeben ist.

Die Bestandteile standen der Behörde in der Sprache: zur Verfügung bzw. wurden in dieser Sprache eingereicht; dabei handelt es sich um

- ☐ die Sprache der Übersetzung, die für die Zwecke der internationalen Recherche eingereicht worden ist (nach Regel 23.1(b)).
- ☐ die Veröffentlichungssprache der internationalen Anmeldung (nach Regel 48.3(b)).
- ☐ die Sprache der Übersetzung, die für die Zwecke der internationalen vorläufigen Prüfung eingereicht worden ist (nach Regel 55.2 und/oder 55.3).

3. Hinsichtlich der in der internationalen Anmeldung offenbarten **Nucleotid- und/oder Aminosäuresequenz** ist die internationale vorläufige Prüfung auf der Grundlage des Sequenzprotokolls durchgeführt worden, das:

- ☐ in der internationalen Anmeldung in schriftlicher Form enthalten ist.
- ☐ zusammen mit der internationalen Anmeldung in computerlesbarer Form eingereicht worden ist.
- ☐ bei der Behörde nachträglich in schriftlicher Form eingereicht worden ist.
- ☐ bei der Behörde nachträglich in computerlesbarer Form eingereicht worden ist.
- ☐ Die Erklärung, daß das nachträglich eingereichte schriftliche Sequenzprotokoll nicht über den Offenbarungsgehalt der internationalen Anmeldung im Anmeldezeitpunkt hinausgeht, wurde vorgelegt.
- ☐ Die Erklärung, daß die in computerlesbarer Form erfassten Informationen dem schriftlichen Sequenzprotokoll entsprechen, wurde vorgelegt.

4. Aufgrund der Änderungen sind folgende Unterlagen fortgefallen:

INTERNATIONALER VORLÄUFIGER PRÜFUNGSBERICHT

Internationales Aktenzeichen PCT/AT00/00167

- ☐ Beschreibung, Seiten:
☐ Ansprüche, Nr.:
☐ Zeichnungen, Blatt:

5. ☐ Dieser Bericht ist ohne Berücksichtigung (von einigen) der Änderungen erstellt worden, da diese aus den angegebenen Gründen nach Auffassung der Behörde über den Offenbarungsgehalt in der ursprünglich eingereichten Fassung hinausgehen (Regel 70.2(c)).

(Auf Ersatzblätter, die solche Änderungen enthalten, ist unter Punkt 1 hinzuweisen; sie sind diesem Bericht beizufügen).

6. Etwaige zusätzliche Bemerkungen:

V. Begründete Feststellung nach Artikel 35(2) hinsichtlich der Neuheit, der erfinderischen Tätigkeit und der gewerblichen Anwendbarkeit; Unterlagen und Erklärungen zur Stützung dieser Feststellung

1. Feststellung

Neuheit (N)	Ja: Ansprüche	1-28
	Nein: Ansprüche	
Erfinderische Tätigkeit (ET)	Ja: Ansprüche	1-28
	Nein: Ansprüche	
Gewerbliche Anwendbarkeit (GA)	Ja: Ansprüche	1-28
	Nein: Ansprüche	

2. Unterlagen und Erklärungen
siehe Beiblatt

VII. Bestimmte Mängel der internationalen Anmeldung

Es wurde festgestellt, daß die internationale Anmeldung nach Form oder Inhalt folgende Mängel aufweist:
siehe Beiblatt

Zu Punkt V

Die Anmeldung betrifft eine elektrische Maschine, vorzugsweise in Drehstromausführung in der Kraftfahrzeugtechnik.

Als nächstkommender Stand der Technik wird D1 (EP-A-769403) angesehen.

Aus der D1 ist ein Hybrid-Antriebssystem bekannt (siehe Abbildungen 2,8), bei dem eine erste (16) und eine zweite (22) elektrische Maschine vorgesehen sind. Die erste elektrische Maschine ist über ihren Rotor mit einer rotierenden Welle einer Verbrennungskraftmaschine (12) mechanisch verbunden. Die zweite elektrische Maschine ist mit ihrem Rotor mit einem rotierenden Teil eines mechanischen Aggregates, in Form eines Getriebes (20), mechanisch gekuppelt.

Der Gegenstand des Anspruchs 1 unterscheidet sich von der Maschinen-anordnung des Dokuments D1 dadurch, dass:

die erste elektrische Maschine mit mindestens der zweiten elektrischen Maschine über leistungselektronische Elemente (12,13) zum **Austausch** elektrischer Energie auf **frei wählbarem** Spannungsniveau **elektrisch verbunden** ist.

Der Gegenstand des Anspruchs 1 ist somit neu.

Die unterscheidenden Merkmale ermöglichen einen eigenen, autarken, internen elektrischen Kreis, der unabhängig vom Spannungsniveau des Bordnetzes ist.

Die mit vorliegender Anmeldung zu lösende **Aufgabe** kann somit in der Angabe einer elektrischen Maschinen-anordnung gesehen werden, bei der der interne elektrische Kreis und damit das Spannungsniveau der ersten Maschine unabhängig von dem Spannungsniveau eines externen, elektrischen Kreises, des sogenannten Bordnetzes (14 auf Abbildung 2) ist.

Die im neuen Anspruch 1 (der sich aus dem ursprünglichen Anspruch 1 und dem auf Seite 11, Zeilen 4,5 dargestellten Merkmal zusammensetzt) vorgeschlagene **Lösung** ist neu und erfinderisch weil sich im Stand der Technik insgesamt keine Lehre findet, die den mit dem technischen Problem befassten Fachmann veranlassen würde, den nächstliegenden Stand der Technik (in dem beide Maschinen an das Batterienspannungsniveau gebunden sind) unter Berücksichtigung dieser Lehre zu ändern oder anzupassen und somit zu etwas zu gelangen, was unter den Patentanspruch fällt (d.h Maschinen die **aneinander elektrisch gekoppelt sind**), und das zu erreichen, was mit der Erfindung erreicht wird (**d.h Austausch** elektrischer Energie auf **frei wählbarem** Spannungsniveau).

In EP-A-0725474 und EP-A0800951 sind auch beide Maschinen an das Batterienspannungsniveau gebunden.

Der Gegenstand des unabhängigen Anspruchs 1 erfüllt somit die Erfordernisse der Artikel 33(2) , 33(3) PCT .

Die Ansprüche 2-28 sind abhängige Ansprüche, die im Zusammenhang mit dem Anspruch 1 die Erfordernisse der Artikel 33(2), 33(3) PCT erfüllen.

Die gewerbliche Anwendbarkeit der Gegenstände der Ansprüche 1-28 steht außer Zweifel. Damit erfüllen die Ansprüche 1-28 auch die Erfordernisse des Art. 33 (4) PCT.

Zu Punkt VII

1. Im Widerspruch zu den Erfordernissen der Regel 5.1 a) ii) PCT werden in der Beschreibung weder der in dem Dokument D1 offenbarte einschlägige Stand der Technik noch dieses Dokument angegeben.

2. a) Die Beschreibung steht nicht, wie in Regel 5.1 a) iii) PCT vorgeschrieben, in Einklang mit den Ansprüchen.

03. 08. 2001

Neue Patentansprüche:

(75)

1. Elektrische Maschine, vorzugsweise in Drehstromausführung, wobei eine
erste elektrische Maschine (10) vorgesehen ist, die über ihren Rotor (5) mit
5 einer rotierenden Welle einer Maschine, insbesondere einer
Verbrennungskraftmaschine, mechanisch verbunden ist und mindestens
eine zweite elektrische Maschine (11) vorgesehen ist, die mit ihrem Rotor
(6) mit einem rotierenden Teil eines mechanischen Aggregates mechanisch
gekuppelt ist, dadurch gekennzeichnet, daß die erste elektrische Maschine
10 (10) mit mindestens der zweiten elektrischen Maschine (11) über
leistungselektronische Elemente zum Austausch elektrischer Energie auf frei
wählbarem Spannungsniveau elektrisch verbunden ist.
2. Elektrische Maschine nach Anspruch 1, dadurch gekennzeichnet, daß die
15 zweite elektrische Maschine (11) mit ihrem Rotor (6) mit dem rotierenden
Teil einer Strömungsmaschine verbunden ist.
3. Elektrische Maschine nach Anspruch 1 oder 2, dadurch gekennzeichnet,
daß die erste elektrische Maschine (10) mit ihrem Rotor (5) mit einer
20 Kurbelwelle oder einer mit der Kurbelwelle in mechanischer Verbindung
stehenden Welle einer Verbrennungskraftmaschine mechanisch verbunden
ist.
4. Elektrische Maschine nach Anspruch 1 oder 2, dadurch gekennzeichnet,
25 daß die erste elektrische Maschine (10) mit der Verbrennungskraftmaschine
über ein Getriebe mechanisch verbunden ist.
5. Elektrische Maschine nach Anspruch 1 oder 2, dadurch gekennzeichnet,
daß die erste elektrische Maschine (10) ein Teil der
30 Verbrennungskraftmaschine ist, beispielsweise daß der Rotor (5) der ersten
elektrischen Maschine (10) in die Schwungscheibe der
Verbrennungskraftmaschine integriert ist.

6. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 5, dadurch gekennzeichnet, daß die erste elektrische Maschine (10) mit mindestens einem externen elektrischen Kreis, vorzugsweise einem Bordnetz (14), verbunden ist.

5

7. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 6, dadurch gekennzeichnet, daß die erste (10) und die zweite elektrische Maschine (11) in einem Gehäuse (9) angeordnet sind.

10 8. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 7, dadurch gekennzeichnet, daß die erste (10) und/oder die zweite elektrische Maschine (11) als Asynchron-, Synchron- oder Reluktanzmaschine ausgeführt ist bzw. sind.

15 9. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 8, dadurch gekennzeichnet, daß die erste (10) und die zweite elektrische Maschine (11) Rotoren (5, 6) mit gleicher Rotationsachse aufweisen.

20 10. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 9, dadurch gekennzeichnet, daß eine der beiden Maschinen (10, 11) als Innenläufer und die andere Maschine als Außenläufer ausgeführt sind.

25 11. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 10, dadurch gekennzeichnet, daß die beiden elektrischen Maschinen (10, 11) ein gemeinsames Statorblechpaket aufweisen.

30 12. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 11, dadurch gekennzeichnet, daß die Komponenten für den elektrischen Energieaustausch zwischen den elektrischen Maschinen (10, 11) und/oder einem externen elektrischen Kreis (14) in einem Gehäuse (9) mindestens einer elektrischen Maschine (10, 11) angeordnet sind.

13. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 12,
dadurch gekennzeichnet, daß das Gehäuse (9) mindestens einer
elektrischen Maschine (10, 11) eine Flüssigkeitskühlung aufweist.
- 5 14. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 13,
dadurch gekennzeichnet, daß vom elektrischen Kreis, der die beiden
elektrischen Maschinen verbindet, ein Netzanschluß mit Gleich-, Wechsel-
oder Drehspannung ableitbar ist.
- 10 15. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 14,
dadurch gekennzeichnet, daß der Stator (1, 4) mindestens einer
elektrischen Maschine (10, 11) mindestens zwei, vorzugsweise in der
Maschine (10, 11) galvanisch getrennte, Wicklungssysteme (22, 23)
aufweist, die mit dem Hauptfluß der Maschine (10, 11) magnetisch
15 gekoppelt sind.
16. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 15,
dadurch gekennzeichnet, daß die mindestens zwei Wicklungssysteme (22,
23) über getrennte leistungselektronische Schaltungen (24, 25) mit
20 jeweiligen, vorzugsweise galvanisch getrennten, Stromkreisen verbunden
sind.
17. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 16,
dadurch gekennzeichnet, daß mindestens ein Wicklungssystem (22, 23)
25 über eine Gleichrichterbrücke mit einem Gleichspannungs- oder
batteriegestützten Netz, vorzugsweise einem Bordnetz (26), zum
Energieaustausch in einer Richtung verbunden ist.
18. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 17,
30 dadurch gekennzeichnet, daß mindestens ein Wicklungssystem (22, 23)
über eine Transistorbrücke mit einem Gleichspannungs- oder


batteriegestützten Netz, vorzugsweise einem Bordnetz (26), zum Energieaustausch in beiden Richtungen verbunden ist.

19. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 18,
5 dadurch gekennzeichnet, daß mit mindestens einem der Wicklungssysteme (22, 23) die Maschine als Generator zum Laden des angeschlossenen Bordnetzes (26), sowie auch als Motor, vorzugsweise als Starter einer mechanisch gekoppelten Verbrennungskraftmaschine, betreibbar ist.
- 10 20. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 19, dadurch gekennzeichnet, daß über die mindestens zwei Wicklungssysteme (22, 23) ein galvanisch trennbarer elektrischer Energieaustausch zwischen den an die Wicklungssysteme (22, 23) angeschlossenen Stromkreisen durchführbar ist.
- 15 21. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 20, dadurch gekennzeichnet, daß die über leistungselektronische steuerbare Schalter angesteuerten Wicklungssysteme (22, 23) die Führung der elektrischen Größen von über leistungselektronische, nicht steuerbare
20 Elemente, vorzugsweise Dioden, gekoppelte Wicklungssysteme (22, 23) übernehmen.
22. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 21, dadurch gekennzeichnet, daß jedes Wicklungssystem (22, 23) galvanisch
25 unabhängig vom jeweiligen anderen Wicklungssystem (22, 23) mit elektromechanischen Funktionsgruppen auf im allgemeinen unterschiedlichen Spannungsebenen verbunden ist.
23. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 22,
30 dadurch gekennzeichnet, daß durch enge magnetische Koppelung von den Wicklungssystemen (22, 23) ein elektromagnetischer Energieaustausch

zwischen diesen Wicklungssystemen (22, 23) unabhängig von einer Rotordrehung nach dem Prinzip des Transformators gegeben ist.

24. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 23,
5 dadurch gekennzeichnet, daß durch schwache magnetische Koppelung von den Wicklungssystemen (22, 23) eine geringe elektromagnetische Beeinflussung der Wicklungssysteme (22, 23) erfolgt.
25. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 24,
10 dadurch gekennzeichnet, daß durch Steuerung der elektromagnetischen Größen, vorzugsweise der Ströme und Flussverkettungen, mindestens eines Wicklungssystems (22, 23) ein beliebig gestaltbarer elektromechanischer Energieaustausch zwischen den Wicklungssystemen (22, 23) und der Rotorwelle erreichbar ist.
- 15 26. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 25, dadurch gekennzeichnet, daß eine erste und die zweite elektrische Maschine (10, 11) in einem Gehäuse angeordnet sind.
- 20 27. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 26, dadurch gekennzeichnet, daß die erste und/oder die zweite elektrische Maschine (10, 11) als Asynchron-, Synchron- oder Reluktanzmaschine ausgeführt ist bzw. sind.
- 25 28. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 27, dadurch gekennzeichnet, daß die erste und die zweite elektrische Maschine (10, 11) Rotoren mit gleicher Rotationsachse aufweisen.

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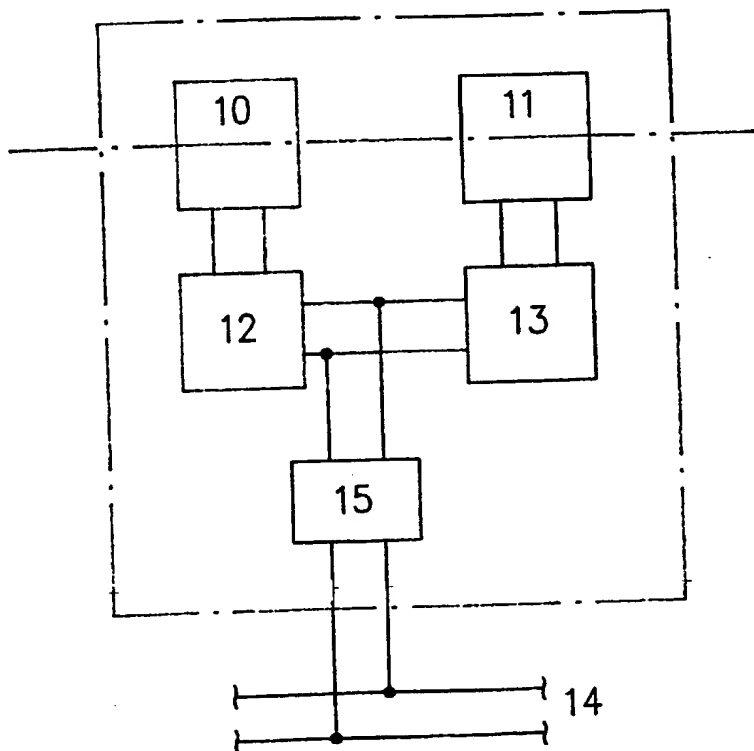
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(54) Title: ELECTRIC MOTOR

(54) Bezeichnung: ELEKTRISCHE MASCHINE



(57) Abstract: The invention relates to an electric motor, preferably of the three-phase current design. According to the invention, a first electric motor (10) is provided which is mechanically connected via the rotor (5) thereof to a rotating shaft of an engine, especially of an internal combustion engine. In addition, at least one second electric motor (11) is provided, whereby the second electric motor (11) is mechanically coupled via the rotor (6) thereof to a rotating part of a mechanical aggregate, especially to a turbo-engine. The first electric motor (10) is electrically coupled to at least the second electric motor (11) in order to exchange electrical power at a freely selectable voltage level.

(57) Zusammenfassung: Die Erfindung betrifft eine elektrische Maschine, vorzugsweise in Drehsstromausführung. Es ist eine erste elektrische Maschine (10) vorgesehen, die über ihren Rotor (5) mit einer rotierenden Welle einer Maschine, insbesondere einer Verbrennungskraftmaschine, mechanisch verbunden ist. Ferner ist mindestens eine zweite elektrische Maschine (11) vorgesehen, wobei

die zweite elektrische Maschine (11) mit ihrem Rotor (6) mit einem rotierenden Teil eines mechanischen

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Zur Erklärung der Zweibuchstaben-Codes, und der anderen Abkürzungen wird auf die Erklärungen ("Guidance Notes on Codes and Abbreviations") am Anfang jeder regulären Ausgabe der PCT-Gazette verwiesen.

Aggregates, insbesondere einer Strömungsmaschine, mechanisch gekuppelt ist. Die erste elektrische Maschine (10) ist mit mindestens der zweiten elektrischen Maschine (11), zum Austausch elektrischer Energie auf frei wählbarem Spannungsniveau, elektrisch verbunden.

Elektrische Maschine

Die Erfindung betrifft eine elektrische Maschine, vorzugsweise in Drehstromausführung.

5

Immer häufiger werden elektrische Maschinen in der Kraftfahrzeugtechnik eingesetzt. So sind Anordnungen, wie beispielsweise das ISAD-System (Integrated Starter-Alternator-Damper System) bekannt, das den Energieaustausch auf Bordspannungsniveau abwickelt.

10

Ferner sind auch elektrisch betriebene Turbolader bekannt, bei denen ebenfalls der Energieaustausch auf Bordspannungsniveau durchgeführt wird. Dabei wird die Turbolader – Leistung zur Gänze aus dem Bordnetz entnommen.

15

Aufgabe der Erfindung ist es, eine elektrische Maschine zu schaffen, die insbesondere in der Kraftfahrzeugtechnik eingesetzt werden kann und die zur Versorgung zweier unterschiedlicher Netze, insbesondere für den Turbolader ausreichende elektrische Energie oder verschiedene Spannungsniveaus zur Verfügung stellt.

20

Die Aufgabe wird durch die Erfindung gelöst. Die erfindungsgemäße elektrische Maschine ist dadurch gekennzeichnet, daß eine erste elektrische Maschine vorgesehen ist, die über ihren Rotor mit einer rotierenden Welle einer Maschine, insbesondere einer Verbrennungskraftmaschine, mechanisch

25

verbunden ist, daß mindestens eine zweite elektrische Maschine vorgesehen ist, daß die zweite elektrische Maschine mit ihrem Rotor mit einem rotierenden Teil eines mechanischen Aggregates, insbesondere einer Strömungsmaschine, mechanisch gekuppelt ist und daß die erste elektrische Maschine mit mindestens der zweiten elektrischen Maschine, zum Austausch elektrischer

30

Energie auf frei wählbarem Spannungsniveau, elektrisch verbunden ist. Mit der Erfindung ist es erstmals möglich einen eigenen, autarken, internen elektrischen Kreis zu schaffen, der unabhängig vom Spannungsniveau des

Bordnetzes ist. Damit können die erfindungsgemäße Maschine und die leistungselektronischen Schaltungen mit optimalen Betriebsspannungen ausgelegt werden. Es ist ja bekannt, daß es üblicherweise vorteilhafter ist, mit höheren Spannungen als denen derzeit üblicher Bordnetze elektrische Leistung zu transportieren.

Dieser interne elektrische Teil ist mit der ersten Maschine über leistungselektronische Elemente wie Dioden und Transistoren gemäß dem Stand der Technik entsprechenden Schaltungen verbunden, um damit den internen elektrischen Teil in seinen elektrischen Kenngrößen, wie Spannungen und Ströme und deren zeitliche Verläufe zu gestalten.

Die erste Maschine kann mit dem Verbrennungsmotor mechanisches Drehmoment zu- und abführen, wodurch die erste Maschine bei Leistungsentnahme generatorisch arbeiten kann und diese Energie in Form elektrischer Energie an den internen elektrischen Teil abgibt. Wenn die erste Maschine vom internen elektrischen Teil Energie bezieht, arbeitet sie als Motor und kann dieses Drehmoment beispielsweise zum Starten der Verbrennungskraftmaschine oder zur Unterstützung oder Optimierung im Betrieb verwenden.

Nach einem besonderen Merkmal der Erfindung ist die erste elektrische Maschine mit ihrem Rotor mit einer Kurbelwelle oder einer mit der Kurbelwelle in mechanischer Verbindung stehenden Welle einer Verbrennungskraftmaschine mechanisch verbunden. Dadurch kann in einfachster Weise das mechanische Drehmoment zwischen der ersten elektrischen Maschine und der Verbrennungskraftmaschine ausgetauscht werden.

Gemäß einer Ausgestaltung der Erfindung ist die erste elektrische Maschine mit der Verbrennungskraftmaschine über ein Getriebe mechanisch verbunden.

Auch diese konstruktive Lösung verbessert bei einem elektrisch angetriebenen Turbolader das Drehmoment bei niedrigen Touren.

5 Nach einer weiteren Ausgestaltung der Erfindung ist die erste elektrische Maschine ein Teil der Verbrennungskraftmaschine, beispielsweise ist der Rotor in der ersten elektrischen Maschine in die Schwungscheibe der Verbrennungskraftmaschine integriert. Der Vorteil dieser konstruktiven Lösung liegt vor allem darin, daß die komplette Anordnung in einer relativ kleinen Bauweise hergestellt werden kann.

10 Nach einem besonderen Merkmal der Erfindung ist die erste elektrische Maschine mit mindestens einem externen elektrischen Kreis, vorzugsweise einem Bordnetz, verbunden. Diese zweite elektrische Koppelstelle ist über eine leistungselektronische Spannungsanpassungsschaltung entsprechend dem Stand
15 der Technik mit dem Bordnetz verbunden. Damit kann Leistung zwischen dem internen elektrischen Teil und dem Bordnetz ausgetauscht werden. Damit kann die erste elektrische Maschine der erfindungsgemäßen Anordnung in der einen Energierichtung als Starter und in der anderen Energierichtung als Bordnetzladeeinrichtung betrieben werden.

20 Gemäß einer weiteren Ausgestaltung der Erfindung sind die erste und die zweite elektrische Maschine in einem Gehäuse angeordnet. Mit dieser Ausgestaltung ist es möglich, ein elektrisches Antriebssystem zu schaffen, das wirtschaftlich hergestellt und eingesetzt werden kann. Vorteilhaft bei dieser
25 Erfindung ist, daß gegenüber bekannten elektromechanischen Antrieben oder rein mechanischen Antrieben, wie Getrieben, bei denen zwei verschiedene, vorzugsweise unabhängige Drehzahlen benötigt werden, wesentliche Teile, wie beispielsweise Gehäuseelemente, Teile der Steuerung eingespart werden können. Weiters können die bekannten EMV-Probleme lokal im Gehäuse gelöst
30 werden und dringen nicht in die Umgebung.

Nach einer Weiterbildung der Erfindung ist bzw. sind die erste und/oder die zweite elektrische Maschine als Asynchron-, Synchron- oder Reluktanzmaschine ausgeführt. Dadurch kann für jeden Anwendungsfall die optimale Maschine gewählt werden.

- 5 Nach einem weiteren Merkmal der Erfindung weisen die erste und die zweite elektrische Maschine Rotoren mit gleicher Rotationsachse auf. Gerade in der Kraftfahrzeugtechnik ist es von Vorteil, wenn für eine mechanisch-elektrisch-mechanische Kupplung nur eine Rotationsachse gegeben ist.
- 10 Gemäß einer besonderen Ausgestaltung der Erfindung sind eine der beiden Maschinen als Innenläufer und die andere Maschine als Außenläufer ausgeführt. Auch durch diese Ausgestaltung der Erfindung ist eine kompakte Ausführung der Maschine möglich.
- 15 Nach einem weiteren Merkmal der Erfindung weisen die beiden elektrischen Maschinen ein gemeinsames Statorblechpaket auf. Bei dieser Ausgestaltung können in einem Gehäuse ein Stator mit mindestens einer Statorwicklung und mindestens zwei Rotoren vorgesehen sein. Die Rotoren sind mechanisch voneinander getrennt und jeder Rotor steht mit dem elektromagnetisch aktiven
- 20 Stator in elektromagnetischer Wechselwirkung, wobei die Drehzahlen der Rotoren gleich oder unterschiedlich sind.

- Gemäß einer Weiterbildung der Erfindung sind die Komponenten für den elektrischen Energieaustausch zwischen den elektrischen Maschinen und/oder
- 25 einem externen elektrischen Kreis in einem Gehäuse mindestens einer elektrischen Maschine angeordnet. Diese Weiterbildung trägt vor allem dazu bei, eine elektrische Maschine für die Kraftfahrzeugtechnik zu schaffen, die eine kompakte Bauweise aufweist.

- 30 Nach einer weiteren Ausgestaltung der Erfindung weist das Gehäuse mindestens einer elektrischen Maschine eine Flüssigkeitskühlung auf. Dadurch kann die Verlustwärme der Wicklungen, aber auch der leistungselektronischen

Elemente, die auf Grund der bekannten Probleme mit den hohen Strömen in der Maschine auftreten kann, optimal abgeführt werden.

Gemäß einem weiteren Merkmal der Erfindung ist vom elektrischen Kreis, der die beiden elektrischen Maschinen verbindet, ein Netzanschluß mit Gleich-, Wechsel- oder Drehspannung ableitbar. In einer dieser Ausgestaltung kann vom internen elektrischen Kreis ein weiteres Dreh-, Wechsel- oder

Gleichspannungsnetz zur Verfügung gestellt werden. Beispielsweise kann ein kräftiges 230 V-Netz oder 3x400 V-Netz ausgekoppelt werden, wobei die

Frequenz entweder intern oder extern vorgegeben werden kann. Damit wird das Bordnetz und die an ihm angeschlossenen Aggregate über den internen elektrischen Kreis mit diesem Netz energiemäßig verbunden.

Damit kann beispielsweise die Verbrennungskraftmaschine vom Netz gestartet werden, ohne das Bordnetz zu benötigen oder umgekehrt die

Verbrennungskraftmaschine ein bestehendes Netz stützen oder aufbauen. Es kann auch die Bordnetzatterie in einfacher Weise vom Netz geladen werden.

Nach einem besonderen Merkmal der Erfindung weist der Stator mindestens einer elektrischen Maschine mindestens zwei, vorzugsweise in der Maschine galvanisch getrennte, Wicklungssysteme auf, die mit dem Hauptfluß der Maschine magnetisch gekoppelt sind. Mit dieser Ausgestaltung der Erfindung ist es möglich, zwei autarke elektrische Kreise zu schaffen, die ein voneinander unabhängiges Spannungsniveau aufweisen. Ferner ist es ein Vorteil dieser

Erfindung, daß elektro-magnetische, also EMV-Störungen durch das Schalten in einem Wicklungssystem in einem anderen Wicklungssystem unterdrückt werden können. So können weiters auch vorteilhaft die einzelnen

Wicklungssysteme auf verschiedenen Spannungsniveaus, insbesondere galvanisch trennbar, arbeiten. Eine eigene galvanische Trennung und/oder ein Transformator zur Spannungsanpassung zwischen den beiden beteiligten

Stromkreisen wird nicht mehr benötigt.

Gemäß einem besonderen Merkmal der Erfindung sind die mindestens zwei Wicklungssysteme über getrennte leistungselektronische Schaltungen mit jeweiligen, vorzugsweise galvanisch getrennten, Stromkreisen verbunden. Dadurch ist der Vorteil gegeben, daß beispielsweise ein Netz, insbesondere ein Bordnetz, von einem weiteren Netz getrennt betrieben und geregelt werden kann.

Nach einer weiteren Ausgestaltung der Erfindung ist mindestens ein Wicklungssystem über eine Gleichrichterbrücke mit einem Gleichspannungs- oder batteriegestützten Netz, vorzugsweise einem Bordnetz, zum Energieaustausch in einer Richtung verbunden ist. Durch diese Ausgestaltung können wirtschaftlichere, oder auch billigere, leistungselektronische Komponenten zum Laden verwendet werden.

Gemäß einem weiteren Merkmal der Erfindung ist mindestens ein Wicklungssystem über eine Transistorbrücke mit einem Gleichspannungs- oder batteriegestützten Netz, vorzugsweise einem Bordnetz, zum Energieaustausch in beiden Richtungen verbunden. Dadurch ist der Vorteil gegeben, daß auf einen eigenen Starter verzichtet werden kann oder es wird dem einen Netz, vorzugsweise dem Bordnetz Energie entnommen und das andere Netz gespeist.

Nach einem besonderen Merkmal der Erfindung ist mit mindestens einem der Wicklungssysteme die Maschine als Generator zum Laden des angeschlossenen Bordnetzes, sowie auch als Motor, vorzugsweise als Starter einer mechanisch gekoppelten Verbrennungskraftmaschine, betreibbar. Auch bei dieser Ausgestaltung ist der Vorteil gegeben, daß der Starter, aber auch die Lichtmaschine bei der Konzeption in Wegfall geraten können.

Gemäß einer Weiterbildung der Erfindung ist über die mindestens zwei Wicklungssysteme ein galvanisch trennbarer elektrischer Energieaustausch zwischen den an die Wicklungssysteme angeschlossenen Stromkreisen

durchführbar. Dadurch ist vorteilhafterweise eine Trennung des Bordnetzes vom zweiten Netz gegeben, das durchaus eine höhere Spannung aufweisen kann.

- 5 Nach einer weiteren Ausgestaltung der Erfindung übernehmen die über leistungselektronische steuerbare Schalter angesteuerten Wicklungssysteme die Führung der elektrischen Größen von über leistungselektronische, nicht steuerbare Elemente, vorzugsweise Dioden, gekoppelte Wicklungssysteme. Dabei ist vorteilhaft, daß zur Steuerung des Ladevorganges keine eigenen
- 10 steuerbaren Elemente notwendig sind, sondern auf die steuerbaren Elemente des zweiten Netzes zurückgegriffen werden kann.

- Gemäß einem weiteren Merkmal der Erfindung ist jedes Wicklungssystem galvanisch unabhängig vom jeweiligen anderen Wicklungssystem mit elektro-
- 15 mechanischen Funktionsgruppen auf im allgemeinen unterschiedlichen Spannungsebenen verbunden. Dadurch können die elektro-mechanischen Funktionsgruppen, wie eine elektrisch betriebene Ölpumpe oder Wasserpumpe, oder auch eine elektro-magnetisch betriebene Ventilsteuerung, für Ein- und Auslaßventile bzw. Motorventile, aber auch elektrisch betriebene Lüfter
- 20 unabhängig von der Leistungsbegrenzung der Gleichspannung bzw. der Batterie auf einem vorteilhaften Spannungs- und/oder Stromniveau betrieben werden.

- Nach einer besonderen Ausgestaltung der Erfindung ist durch enge
- 25 magnetische Koppelung von den Wicklungssystemen ein elektromagnetischer Energieaustausch zwischen diesen Wicklungssystemen unabhängig von einer Rotordrehung nach dem Prinzip des Transformators gegeben. Damit ist der Vorteil gegeben, daß auch bei stillstehenden Rotor über eine zeitvariable Spannung durch geeignete leistungselektronische Stellglieder an einem
- 30 Wicklungssystem ein Energietransport in das relativ eng gekoppelte andere Wicklungssystem möglich ist.

Gemäß einem weiteren Merkmal der Erfindung erfolgt durch schwache magnetische Koppelung von den Wicklungssystemen eine geringe elektromagnetische Beeinflussung der Wicklungssysteme. Dadurch ist der Vorteil gegeben, daß elektro-magnetische Störungen durch Schaltvorgänge in einem Wicklungssystem kaum im anderen Wicklungssystem wirksam werden.

Nach einer Weiterbildung der Erfindung ist durch Steuerung der elektromagnetischen Größen, vorzugsweise der Ströme und Flussverkettungen, mindestens eines Wicklungssystems ein beliebig gestaltbarer elektromechanischer Energieaustausch zwischen den Wicklungssystemen und der Rotorwelle erreichbar. Durch diese Ausgestaltung ist der Vorteil gegeben, daß mechanische und elektrische Energie entsprechend der aktuellen, optimalen Strategie zu Verfügung gestellt wird.

Gemäß einer weiteren Ausgestaltung der Erfindung sind eine erste und eine zweite elektrische Maschine in einem Gehäuse angeordnet. Mit dieser Ausgestaltung ist es möglich, ein elektrisches Antriebssystem zu schaffen, das wirtschaftlich hergestellt und eingesetzt werden kann. Vorteilhaft bei dieser Erfindung ist, daß gegenüber bekannten elektromechanischen Antrieben oder rein mechanischen Antrieben, wie Getrieben, bei denen zwei verschiedene, vorzugsweise unabhängige Drehzahlen benötigt werden, wesentliche Teile, wie beispielsweise Gehäuseelemente, Teile der Steuerung eingespart werden können. Weiters können die bekannten EMV-Probleme lokal im Gehäuse gelöst werden und dringen nicht in die Umgebung.

Nach einer Weiterbildung der Erfindung ist bzw. sind die erste und/oder die zweite elektrische Maschine als Asynchron-, Synchron- oder Reluktanzmaschine ausgeführt. Dadurch kann für jeden Anwendungsfall die optimale Maschine gewählt werden.

Nach einem weiteren Merkmal der Erfindung weisen die erste und die zweite elektrische Maschine Rotoren mit gleicher Rotationsachse auf. Gerade in der

Kraftfahrzeugtechnik ist es von Vorteil, wenn für eine mechanisch-elektrisch-mechanische Kupplung nur eine Rotationsachse gegeben ist.

Die Erfindung wird an Hand von Ausführungsbeispielen, die in der Zeichnung
5 dargestellt sind, näher erläutert.

Fig.1 zeigt eine elektrische Maschine mit Rotoren mit gleicher Rotorachse,

Fig. 2 eine Prinzipskizze der elektrischen Schaltung der Maschine,

10

Fig. 3 die elektrische Maschine mit den elektronischen Elementen,

Fig. 4 und 5 eine Ausführungsvariante der elektrischen Maschine,

15 Fig. 6 eine Prinzipskizze einer elektrischen Schaltung der Maschine und

Fig. 7 eine Zusammenschaltung eines Generators mit einem Verdichtermotor
über Umrichter.

20 Einführend sei festgehalten, daß in der beschriebenen Ausführungsform gleiche
Teile bzw. Zustände mit gleichen Bezugszeichen bzw. gleichen
Bauteilbezeichnungen versehen werden, wobei die in der gesamten
Beschreibung enthaltenen Offenbarungen sinngemäß auf gleiche Teile bzw.
Zustände mit gleichen Bezugszeichen bzw. gleichen Bauteilbezeichnungen
25 übertragen werden können. Auch sind die in der Beschreibung gewählten
Lageangaben, wie z.B. oben, unten, seitlich usw. auf die unmittelbar
beschriebene sowie dargestellte Figur bezogen und sind bei einer
Lageänderung sinngemäß auf die neue Lage zu übertragen.

30 Weiters können auch Einzelmerkmale oder Merkmalskombinationen aus dem
gezeigten Ausführungsbeispielen für sich eigenständige, erfindungsgemäße

Lösungen darstellen. Die diesbezüglichen erfindungsgemäßen Aufgaben und Lösungen sind den detaillierten Beschreibungen dieser Figuren zu entnehmen.

Grundsätzlich sind verschiedene Varianten von derartigen elektrischen Maschinen möglich. Gemäß der Fig. 1 ist eine erste elektrische Maschine 10 mit einem Stator 1 der eine Wicklungen 2 aufweist, dargestellt. Bei der zylinderischen Motoranordnung befindet sich eine Wicklung 2 an der Innenseite des Stators 1 bzw. der Statorbohrung und kann als Nut- oder Luftspaltwicklung ausgeführt sein. Die zweite elektrische Maschine 11 weist eine Wicklung 3 an der Außenseite des Stators 4 als Nut- oder Luftspaltwicklung auf, wobei die Wicklung 2 mit einem als Innenläufer ausgeführten Rotor 5 sowie die Wicklung 3 mit einem als Außenläufer ausgebildeten Rotor 6 zusammenarbeitet. Die Rotoren 5, 6 können mit Permanentmagnetenerregung, als Käfigläufer, im Reluktanzaufbau, etc. ausgeführt sein. Die beiden Rotoren 5, 6 sind mechanisch über je eine geeignete Lagerung 7, 8 nach dem Stand der Technik im Gehäuse 9 gelagert.

Wie bereits erwähnt, soll das Haupteinsatzgebiet einer derartigen elektrischen Maschine 10, 11 in der Kraftfahrzeugtechnik liegen, wobei diese mehrere Funktionen erfüllen kann. So ist die erste elektrische Maschine 10 mit der Verbrennungskraftmaschine mechanisch gekuppelt, z.B. über ein Getriebe mit der Kurbelwelle oder die erste Maschine 10 befindet sich mit deren Rotor 5 direkt auf einem bestehenden Element des Verbrennungsmotors, wie etwa auf der Kupplungsschwungscheibe oder einem vorhandenen Abtriebsrad oder ist konstruktiv in diesen Teil integriert. Diese erste Maschine 10 kann mit dem Verbrennungsmotor daher mechanisches Drehmoment zu- und abführen, wodurch die erste Maschine 10 bei Leistungsentnahme generatorisch arbeiten kann und diese Energie in Form elektrischer Energie an den internen elektrischen Teil abgibt. Wenn die erste Maschine 10 vom internen elektrischen Teil Energie bezieht, arbeitet sie als Motor und kann dieses Drehmoment zum Starten der Verbrennungskraftmaschine oder zur Unterstützung oder Optimierung im Betrieb verwenden.

Gemäß der Fig. 2 ist die erste elektrische Maschine 10 sowie die zweite elektrische Maschine 11 jeweils mit einem Steuer- oder Leistungsteil 12, 13 verbunden. Zum Austausch elektrischer Energie auf frei wählbaren Spannungsniveau sind die beiden Steuer- und Leistungsteile 12, 13, die auch die elektronische Leistungsumformung durchführen miteinander verbunden. Dieser interne elektrische Kreis ist mit der ersten Maschine über leistungselektronische Elemente, wie Dioden und Transistoren in dem Stand der Technik entsprechenden Schaltungen verbunden, um damit den internen elektrischen Teil in seinen elektrischen Kenngrößen, wie Spannungen und Ströme und deren zeitliche Verläufe zu gestalten. Ein wichtiges Merkmal dieses internen elektrischen Kreises und damit des Spannungsniveaus der ersten Maschine ist die Unabhängigkeit des Spannungsniveaus von einem externen, elektrischen Kreis, dem sogenannten Bordnetz 14. Damit können die erfindungsgemäßen Maschinen und leistungselektronischen Schaltungen mit optimalen Betriebsspannungen ausgelegt werden. Es ist bekannt, daß es üblicherweise vorteilhafter ist, mit höheren Spannungen als denen derzeit üblicher Bordnetze elektrische Leistung zu transportieren.

Dieses externe Bordnetz 14 ist über einen weiteren Steuer- bzw. Leistungsteil 15 mit dem internen elektrischen Kreis verbunden.

Darin ist auch ein wesentlicher Vorteil dieser Anordnung gegeben, da der mechanischen Energieaustausch zwischen Verbrennungsmotor und einem oder mehreren weiteren Aggregaten wie Turboladern, Pumpen, Lüftern, Kompressoren etc. ohne Verwendung des Bordnetzes abgewickelt werden kann. Neben dem optimalen Spannungsniveau bietet die Anordnung auch ein wesentlich besseres EMV-Verhalten, da die EMV-Störungen durch einfache Maßnahmen entsprechend dem Stand der Technik nicht an das Bordnetz 14 bzw. allgemein in die Umgebung des Aggregats gelangen, sondern nur innerhalb des Aggregats bewältigt werden müssen. Weiters kann mit der Anordnung wesentlich mehr Energie zur Speisung von Hilfsaggregaten als über das Bordnetz drehzahlunabhängig übertragen werden.

Von diesem internen elektrischen Teil, dessen Spannung an den optimalen Betrieb der Anordnung laufend angepaßt werden kann, sofern die leistungselektronischen Elemente dies ermöglichen, gehen nun eine oder vorzugsweise zwei oder mehrere elektrische Leistungsaustausch-Koppelstellen aus.

Die erste elektrische Koppelstelle geht über leistungselektronische Elemente zum elektrischen Anschluß der zweiten Maschine 11, die elektrische Leistung auf von der ersten Maschine 10 grundsätzlich unabhängigem Drehzahlniveau, in mechanische Leistung umwandeln kann. Diese mechanische Leistung dient in einer bevorzugten Variante dieser Anordnung dazu, eine Strömungsmaschine, wie beispielsweise einen Turbolader zu betreiben, um damit den Vorteil eines verbrennungsmotordrehzahlunabhängigen Betriebes der Strömungsmaschine zu ermöglichen. Gegenüber bekannten elektrisch betriebenen Turboladern weist diese Anordnung ferner den großen Vorteil auf, daß die Turbolader-Leistung nicht notwendigerweise zur Gänze aus dem Bordnetz 14 entnommen wird, sondern ganz oder teilweise über die erste Maschine 10 mit dem Verbrennungsmotor ausgetauscht wird. Dies entlastet das Bordnetz massiv und erlaubt Energieaustausch auf günstigerem elektrischen Spannungsniveau, wodurch die Verkabelung und die leistungselektronischen Komponenten vorteilhafter ausgeführt werden können. Der Leistungsaustausch ist bidirektional möglich. In gleicher Art können weitere elektrische Maschinen als Bestandteil der Erfindung zum Antrieb weiterer Aggregate wie Wasserpumpen, Lüfter, Kompressoren usw. am internen elektrischen Teil angekoppelt sein.

Die zweite elektrische Koppelstelle ist über eine leistungselektronische Spannungsanpaßschaltung entsprechend dem Stand der Technik mit dem Bordnetz verbunden. Damit kann Leistung zwischen dem internen elektrischen Teil und dem Bordnetz 14 ausgetauscht werden. Damit kann die erste elektrische Maschine 10 der Anordnung in der einen Energierichtung als Starter und in der anderen Energierichtung als Bordnetzladeeinrichtung betrieben

- werden. Der große Vorteil dieses Betriebsmodus ist, daß kein eigener Starter und keine eigene Lichtmaschine nötig sind, weil diese Funktionen von der Anordnung abgedeckt werden. Ein wesentlicher Vorteil gegenüber bekannten Anordnungen ist, daß der Starter nun als Maschine mit optimalem
- 5 Spannungsniveau ausgelegt und betrieben werden kann, so daß die bekannten Probleme mit hohen Strömen in der Maschine und auch in der an den Maschinensträngen angeschlossenen leistungselektronischen Elementen vermieden werden.
- 10 Weiters kann die zweite Maschine 11 über das Bordnetz 14 unabhängig vom Verbrennungsmotor, also etwa auch während dessen Stillstand, betrieben werden. Damit kann beispielsweise der Turbolader im Stillstand der Verbrennungskraftmaschine gestartet werden, um einen besseren Startprozess zu ermöglichen. Gegenüber bekannten Lösungen bietet diese Lösung den
- 15 Vorteil, daß der zweite Motor und die motornahe Leistungselektronik für ein optimales Spannungsniveau ausgelegt und betrieben werden kann.

- In einer weiteren Ausgestaltung kann vom internen elektrischen Kreis ein weiteres Dreh-, Wechsel- oder Gleichspannungsnetz zur Verfügung gestellt
- 20 werden. Beispielsweise kann ein kräftiges 230 V-Netz oder 3x400 V-Netz ausgekoppelt werden, wobei die Frequenz entweder intern oder extern vorgegeben werden kann. Damit wird das Bordnetz 14 und die an ihm angeschlossenen Aggregate über den internen elektrischen Kreis mit diesem Netz energiemäßig verbunden.
- 25 Damit kann beispielsweise die Verbrennungskraftmaschine vom Netz gestartet werden, ohne das Bordnetz zu benötigen oder umgekehrt die Verbrennungskraftmaschine ein bestehendes Netz stützen oder aufbauen. Es kann auch die Bordnetzatterie in einfacher Weise vom Netz geladen werden.
- 30 Entsprechend der Fig. 3 ist die erste elektrische Maschine 10 und die zweite elektrische Maschine 11 im Gehäuse 9 angeordnet.

Zur Kühlung kann das Gehäuse 9 Kühlkanäle 16 aufweisen.

Zweckmäßigerweise kann im Bereich dieser gut gekühlten Gehäuseteile eine leistungs- und steuerungselektronische Schaltung einschließlich elektrischer, magnetischer und mechanischer Bauteile, wie Halbleiter 19, Kondensatoren 18, Drosseln, Relais od. dgl. sowie allfälliger Trägermaterialien 17 angeordnet sein, um die Funktionen gemäß den Elementen, wie dem Steuer- und Leistungsteil 12, 13, 15 in Fig. 2, vorteilhaft zu realisieren.

Eine weitere Variante der Bauweise der elektrischen Maschine ist aus der Fig. 4 und der Fig. 5 zu entnehmen. Dabei können die beiden elektrischen Maschinen 10, 11 übereinander angeordnet sein und der Rotorausgang kann bei jeder Maschine links und/oder rechts vorgesehen werden. Dabei kann in diesem Maschinengehäuse auch der elektronische Teil 20 integriert sein.

Gemäß der Fig. 6 ist eine elektrische Maschine in Drehstromausführung dargestellt, wobei diese elektrische Maschine die erste oder die zweite elektrische Maschine sein kann. Der Rotor der ersten elektrischen Maschine ist beispielsweise mit einer rotierenden Welle einer Verbrennungskraftmaschine mechanisch verbunden. Der Rotor der zweiten elektrischen Maschine ist mit einem rotierenden Teil, beispielsweise einer Strömungsmaschine, gekuppelt. Zum Austausch elektrischer Energie auf frei wählbaren Spannungsniveau ist die erste elektrische Maschine mit der zweiten elektrischen Maschine elektrisch verbunden.

Der Stator 21 mindestens einer der beiden elektrischen Maschinen weist mindestens zwei Wicklungssysteme 22 bzw. 23 auf. Die beiden Wicklungssysteme 22, 23 sind in der elektrischen Maschine vorzugsweise galvanisch getrennt und sind mit dem Hauptfluß der Maschine magnetisch gekoppelt. Durch die galvanische Trennung, das heißt jedes Wicklungssystem 22, 23 liegt vorzugsweise in seinen eigenen Nuten, können EMV-Störungen durch das Schalten in einem Wicklungssystem 22, 23 unterdrückt werden.

Die beiden Wicklungssysteme 22, 23 sind über getrennte leistungselektronische Schaltungen 24, 25 mit jeweiligen, ebenfalls vorzugsweise galvanisch getrennten, Stromkreisen verbunden. So kann das Wicklungssystem 22 über die leistungselektronische Schaltung 24, beispielsweise einer

5 Gleichrichterbrücke oder mit einer Transistorbrücke mit einem Gleichspannungs- oder batteriegestützten Netz, vorzugsweise mit dem Bordnetz 26, zum Energieaustausch in einer oder beiden Richtungen verbunden sein. Natürlich könnte dieses Wicklungssystem 22 auch als Motor, vorzugsweise als Starter einer Verbrennungskraftmaschine, betreibbar sein.

10 Über die leistungselektronische Schaltung 25 kann ein Netz 27 gespeist werden. Ebenso ist aber auch diese leistungselektronische Schaltung 25 über das interne Netz mit einem leistungselektronischen Stellglied 28 für die zweite elektrische Maschine 29 elektrisch verbunden sein.

15 Jedes Wicklungssystem 22, 23 ist galvanisch unabhängig vom jeweiligen anderen Wicklungssystem 22, 23 mit elektro-mechanischen Funktionsgruppen auf im allgemeinen unterschiedlichen Spannungsebenen verbunden. Dadurch können die elektro-mechanischen Funktionsgruppen, wie eine elektrisch

20 betriebene Ölpumpe oder Wasserpumpe, oder auch eine elektro-magnetisch betriebene Ventilsteuerung, für Ein- und Auslaßventile bzw. Motorventile, aber auch elektrisch betriebene Lüfter unabhängig von der Leistungsbegrenzung der Gleichspannung bzw. der Batterie auf einem vorteilhaften Spannungs- und/oder Stromniveau betrieben werden.

25 Die Wicklungssysteme 22, 23 können eine schwache magnetische Kopplung aufweisen, beispielsweise wenn die Wicklungssysteme in verschiedenen Nuten untergebracht sind, oder auch eine enge magnetische Kopplung aufweisen, wenn beide Wicklungssysteme 22, 23 in einer Nut angeordnet werden.

30 Gemäß der Fig. 7 ist ein Generator, beispielsweise als erste elektrische Maschine 10 und ein Verdichtermotor als zweite elektrische Maschine 11

aufgezeigt. Die beiden elektrischen Maschinen sind über einen Generator-Umrichter 32 und einem Verdichtermotor-Umrichter 33 elektrisch miteinander verbunden. Mit Uzk ist dabei die Zwischenkreisspannung bezeichnet.

- 5 Der Generator ist mit seinem Rotor mit einer Maschine, insbesondere einer Brennkraftmaschine über ein Getriebe 35 verbunden. Der Verdichtermotor 31 ist mit seinem Rotor mit einer Strömungsmaschine 34 verbunden. Ein Wicklungssystem 22, 23 ist über eine leistungselektronische Schaltung 4 mit einem Bordnetz 6 verbunden, wobei die Wicklungssysteme 22, 23 galvanisch
10 trennbar sind.

Dabei könnte die erste und die zweite elektrische Maschine in einem Gehäuse angeordnet sein. Ebenfalls könnte die erste und die zweite elektrische Maschine Rotoren mit gleicher Rotationsachse aufweisen.

15

Abschließend sei der Ordnung halber darauf hingewiesen, daß in der Zeichnung einzelne Bauteile und Baugruppen zum besseren Verständnis der Erfindung unpropotional und maßstäblich verzerrt dargestellt sind.

20

25

Patentansprüche:

1. Elektrische Maschine, vorzugsweise in Drehstromausführung, dadurch gekennzeichnet, daß eine erste elektrische Maschine (10) vorgesehen ist,
5 die über ihren Rotor (5) mit einer rotierenden Welle einer Maschine, insbesondere einer Verbrennungskraftmaschine, mechanisch verbunden ist, daß mindestens eine zweite elektrische Maschine (11) vorgesehen ist, daß die zweite elektrische Maschine (11) mit ihrem Rotor (6) mit einem rotierenden Teil eines mechanischen Aggregates, insbesondere einer
10 Strömungsmaschine, mechanisch gekuppelt ist und daß die erste elektrische Maschine (10) mit mindestens der zweiten elektrischen Maschine (11), zum Austausch elektrischer Energie auf frei wählbarem Spannungsniveau, elektrisch verbunden ist.
- 15 2. Elektrische Maschine nach Anspruch 1, dadurch gekennzeichnet, daß die erste elektrische Maschine (10) mit ihrem Rotor (5) mit einer Kurbelwelle oder einer mit der Kurbelwelle in mechanischer Verbindung stehenden Welle einer Verbrennungskraftmaschine mechanisch verbunden ist.
- 20 3. Elektrische Maschine nach Anspruch 1, dadurch gekennzeichnet, daß die erste elektrische Maschine (10) mit der Verbrennungskraftmaschine über ein Getriebe mechanisch verbunden ist.
4. Elektrische Maschine nach Anspruch 1, dadurch gekennzeichnet, daß die
25 erste elektrische Maschine (10) ein Teil der Verbrennungskraftmaschine ist, beispielsweise daß der Rotor (5) der ersten elektrischen Maschine (10) in die Schwungscheibe der Verbrennungskraftmaschine integriert ist.
5. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 4,
30 dadurch gekennzeichnet, daß die erste elektrische Maschine (10) mit mindestens einem externen elektrischen Kreis, vorzugsweise einem Bordnetz (14), verbunden ist.

6. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 5, dadurch gekennzeichnet, daß die erste (10) und die zweite elektrische Maschine (11) in einem Gehäuse (9) angeordnet sind.
- 5 7. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 6, dadurch gekennzeichnet, daß die erste (10) und/oder die zweite elektrische Maschine (11) als Asynchron-, Synchron- oder Reluktanzmaschine ausgeführt ist bzw. sind.
- 10 8. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 7, dadurch gekennzeichnet, daß die erste (10) und die zweite elektrische Maschine (11) Rotoren (5, 6) mit gleicher Rotationsachse aufweisen.
- 15 9. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 8, dadurch gekennzeichnet, daß eine der beiden Maschinen (10, 11) als Innenläufer und die andere Maschine als Außenläufer ausgeführt sind.
- 20 10. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 9, dadurch gekennzeichnet, daß die beiden elektrischen Maschinen (10, 11) ein gemeinsames Statorblechpaket aufweisen.
- 25 11. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 10, dadurch gekennzeichnet, daß die Komponenten für den elektrischen Energieaustausch zwischen den elektrischen Maschinen (10, 11) und/oder einem externen elektrischen Kreis (14) in einem Gehäuse (9) mindestens einer elektrischen Maschine (10, 11) angeordnet sind.
- 30 12. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 11, dadurch gekennzeichnet, daß das Gehäuse (9) mindestens einer elektrischen Maschine (10, 11) eine Flüssigkeitskühlung aufweist.

13. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 12, dadurch gekennzeichnet, daß vom elektrischen Kreis, der die beiden elektrischen Maschinen verbindet, ein Netzanschluß mit Gleich-, Wechsel- oder Drehspannung ableitbar ist.

5

14. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 13, dadurch gekennzeichnet, daß der Stator (1, 4) mindestens einer elektrischen Maschine (10, 11) mindestens zwei, vorzugsweise in der Maschine (10, 11) galvanisch getrennte, Wicklungssysteme (22, 23) aufweist, die mit dem Hauptfluß der Maschine (10, 11) magnetisch gekoppelt sind.

10

15. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 14, dadurch gekennzeichnet, daß die mindestens zwei Wicklungssysteme (22, 23) über getrennte leistungselektronische Schaltungen (24, 25) mit jeweiligen, vorzugsweise galvanisch getrennten, Stromkreisen verbunden sind.

15

16. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 15, dadurch gekennzeichnet, daß mindestens ein Wicklungssystem (22, 23) über eine Gleichrichterbrücke mit einem Gleichspannungs- oder batteriegestützten Netz, vorzugsweise einem Bordnetz (26), zum Energieaustausch in einer Richtung verbunden ist.

20

17. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 16, dadurch gekennzeichnet, daß mindestens ein Wicklungssystem (22, 23) über eine Transistorbrücke mit einem Gleichspannungs- oder batteriegestützten Netz, vorzugsweise einem Bordnetz (26), zum Energieaustausch in beiden Richtungen verbunden ist.

25

30

18. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 17, dadurch gekennzeichnet, daß mit mindestens einem der Wicklungssysteme

(22, 23) die Maschine als Generator zum Laden des angeschlossenen Bordnetzes (26), sowie auch als Motor, vorzugsweise als Starter einer mechanisch gekoppelten Verbrennungskraftmaschine, betreibbar ist.

- 5 19. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 18, dadurch gekennzeichnet, daß über die mindestens zwei Wicklungssysteme (22, 23) ein galvanisch trennbarer elektrischer Energieaustausch zwischen den an die Wicklungssysteme (22, 23) angeschlossenen Stromkreisen durchführbar ist.

10

20. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 19, dadurch gekennzeichnet, daß die über leistungselektronische steuerbare Schalter angesteuerten Wicklungssysteme (22, 23) die Führung der elektrischen Größen von über leistungselektronische, nicht steuerbare
15 Elemente, vorzugsweise Dioden, gekoppelte Wicklungssysteme (22, 23) übernehmen.

21. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 20, dadurch gekennzeichnet, daß jedes Wicklungssystem (22, 23) galvanisch
20 unabhängig vom jeweiligen anderen Wicklungssystem (22, 23) mit elektromechanischen Funktionsgruppen auf im allgemeinen unterschiedlichen Spannungsebenen verbunden ist.

22. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 21, dadurch gekennzeichnet, daß durch enge magnetische Koppelung von den
25 Wicklungssystemen (22, 23) ein elektromagnetischer Energieaustausch zwischen diesen Wicklungssystemen (22, 23) unabhängig von einer Rotordrehung nach dem Prinzip des Transformators gegeben ist.

- 30 23. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 22, dadurch gekennzeichnet, daß durch schwache magnetische Koppelung von

den Wicklungssystemen (22, 23) eine geringe elektromagnetische Beeinflussung der Wicklungssysteme (22, 23) erfolgt.

- 5 24. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 23, dadurch gekennzeichnet, daß durch Steuerung der elektromagnetischen Größen, vorzugsweise der Ströme und Flussverkettungen, mindestens eines Wicklungssystems (22, 23) ein beliebig gestaltbarer elektromechanischer Energieaustausch zwischen den Wicklungssystemen (22, 23) und der Rotorwelle erreichbar ist.
- 10 25. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 24, dadurch gekennzeichnet, daß eine erste und die zweite elektrische Maschine (10, 11) in einem Gehäuse angeordnet sind.
- 15 26. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 25, dadurch gekennzeichnet, daß die erste und/oder die zweite elektrische Maschine (10, 11) als Asynchron-, Synchron- oder Reluktanzmaschine ausgeführt ist bzw. sind.
- 20 27. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 26, dadurch gekennzeichnet, daß die erste und die zweite elektrische Maschine (10, 11) Rotoren mit gleicher Rotationsachse aufweisen.

Fig. 1

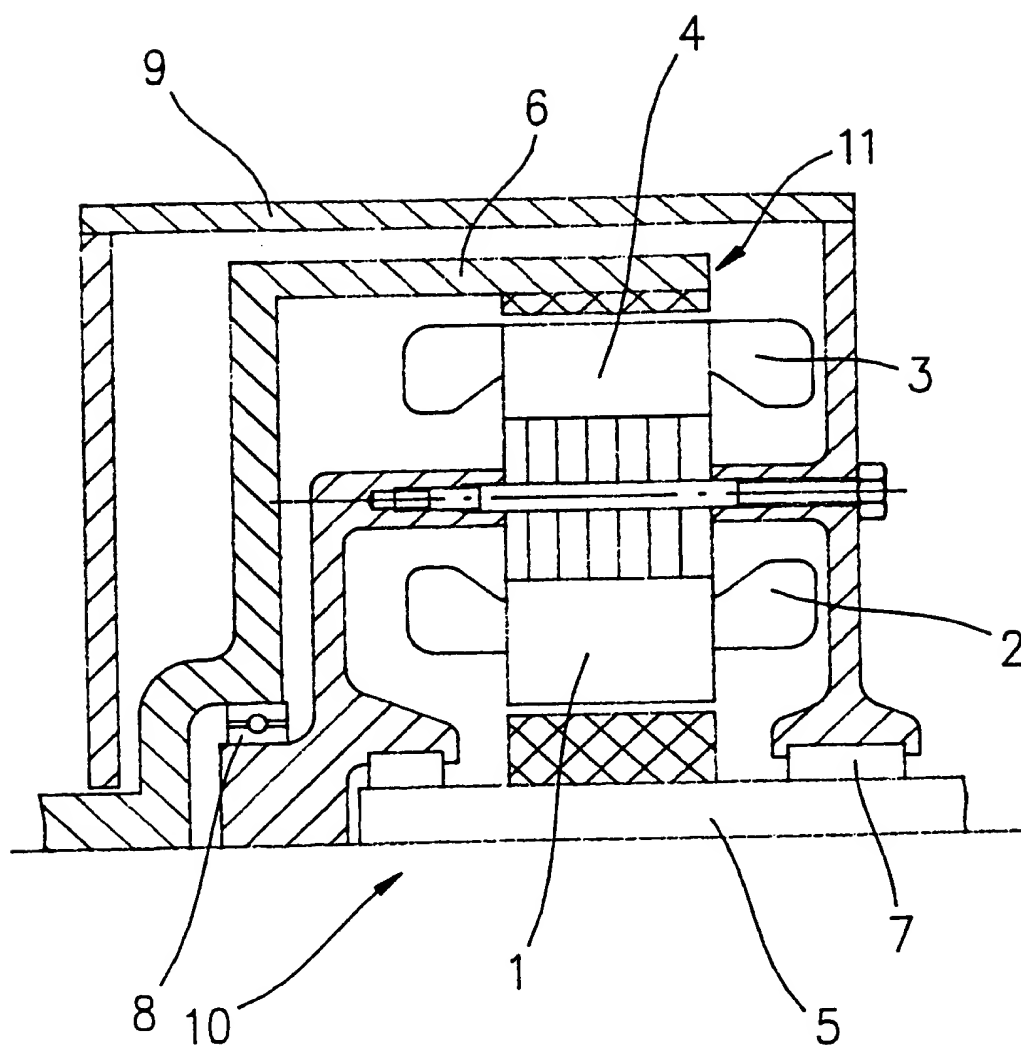


Fig. 2

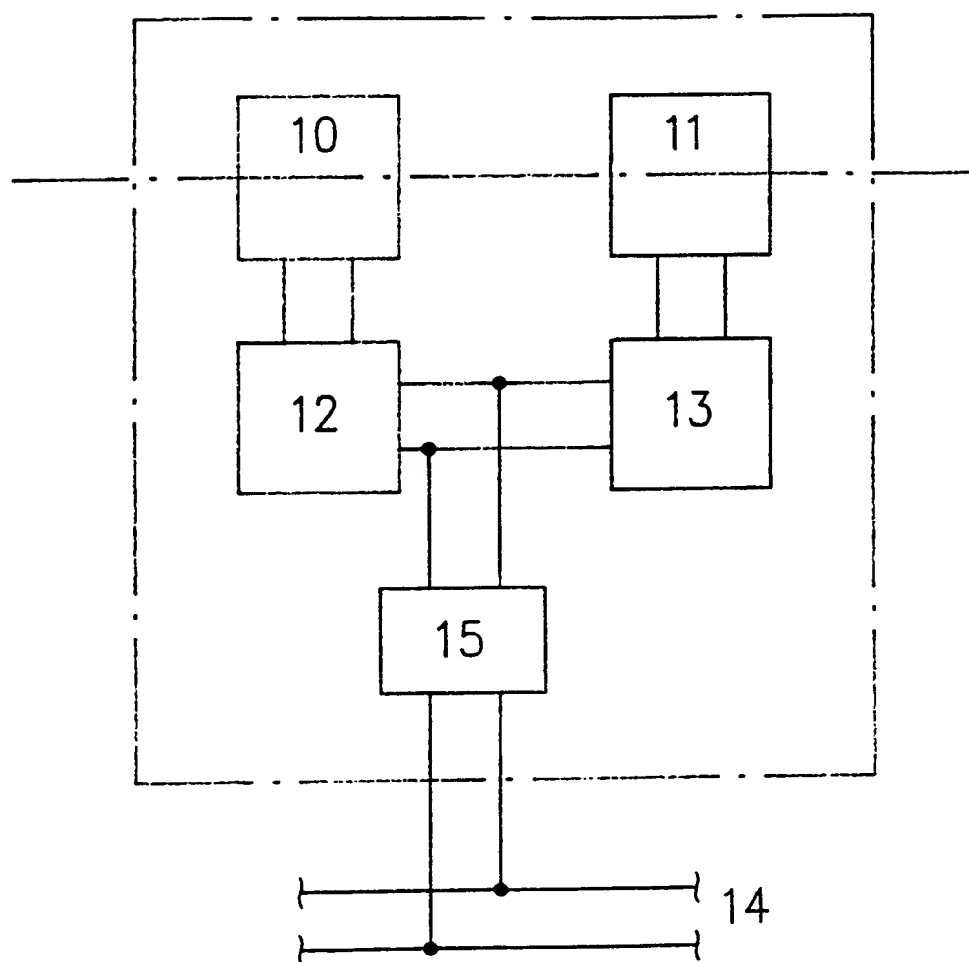


Fig. 3

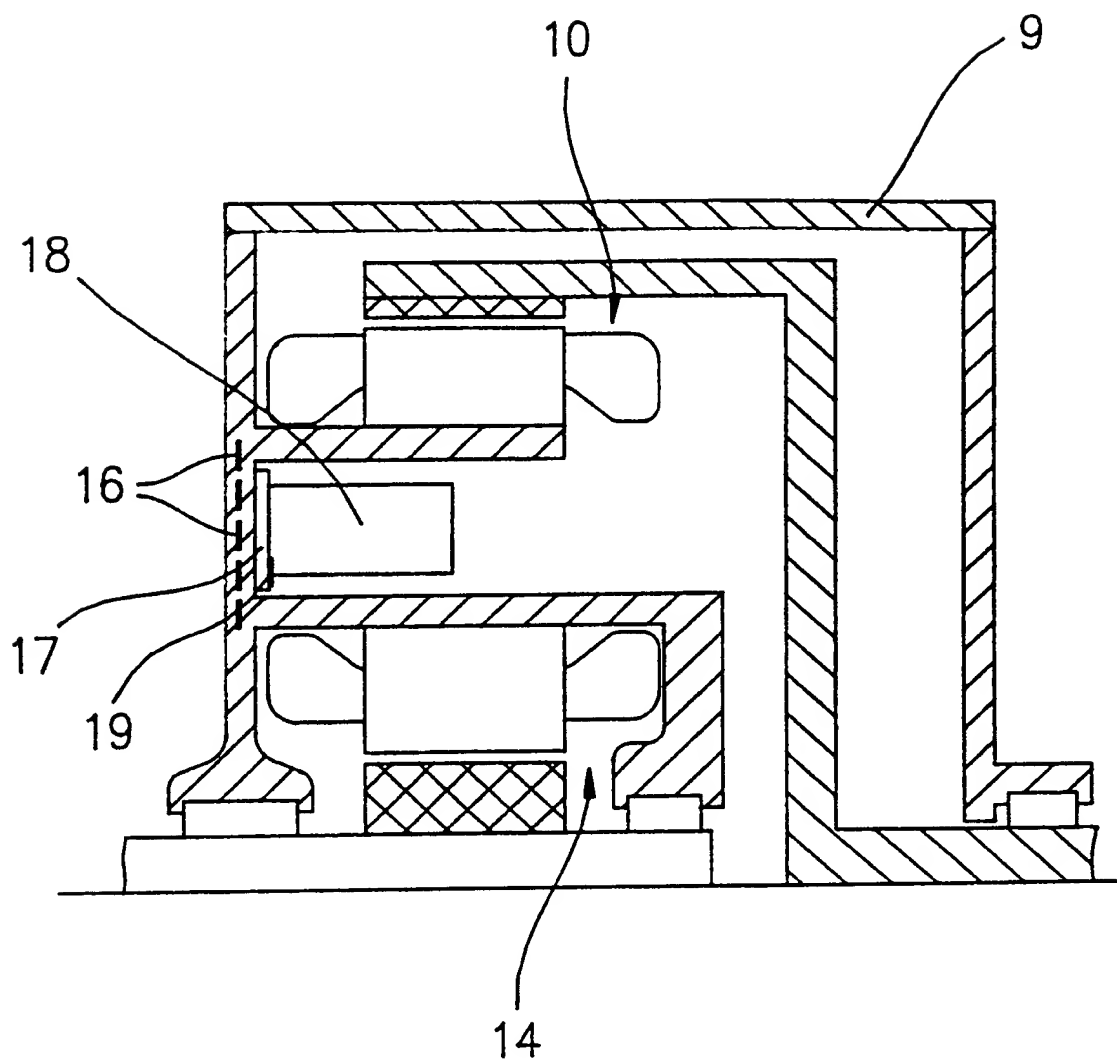


Fig. 4

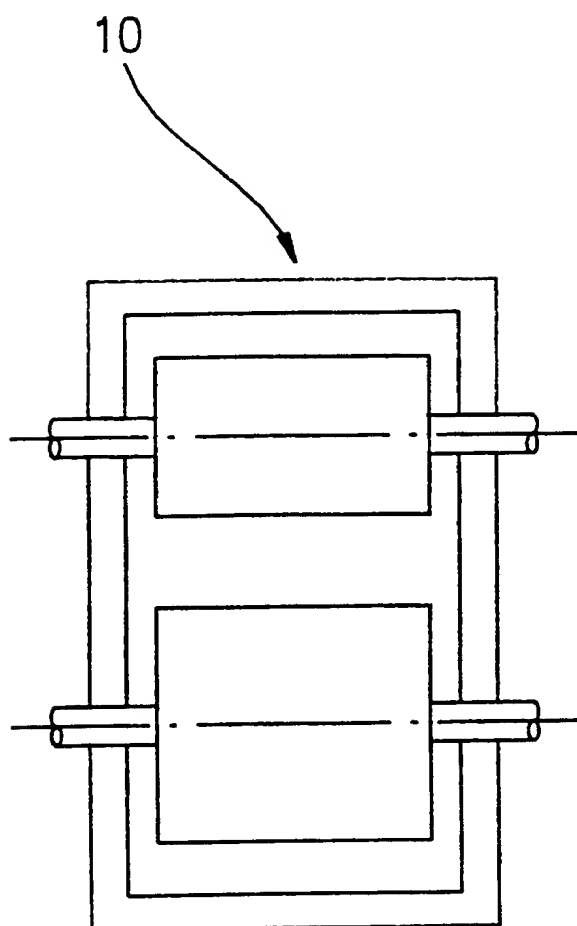
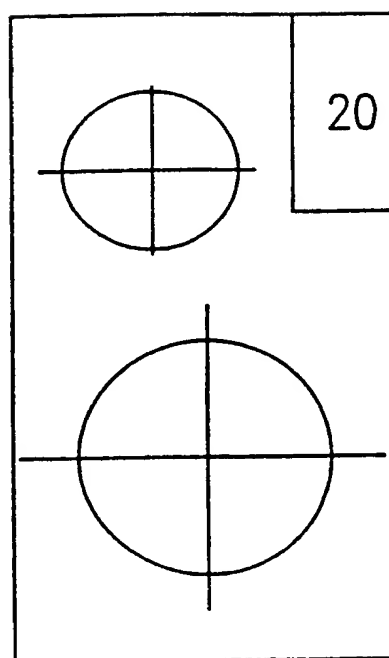
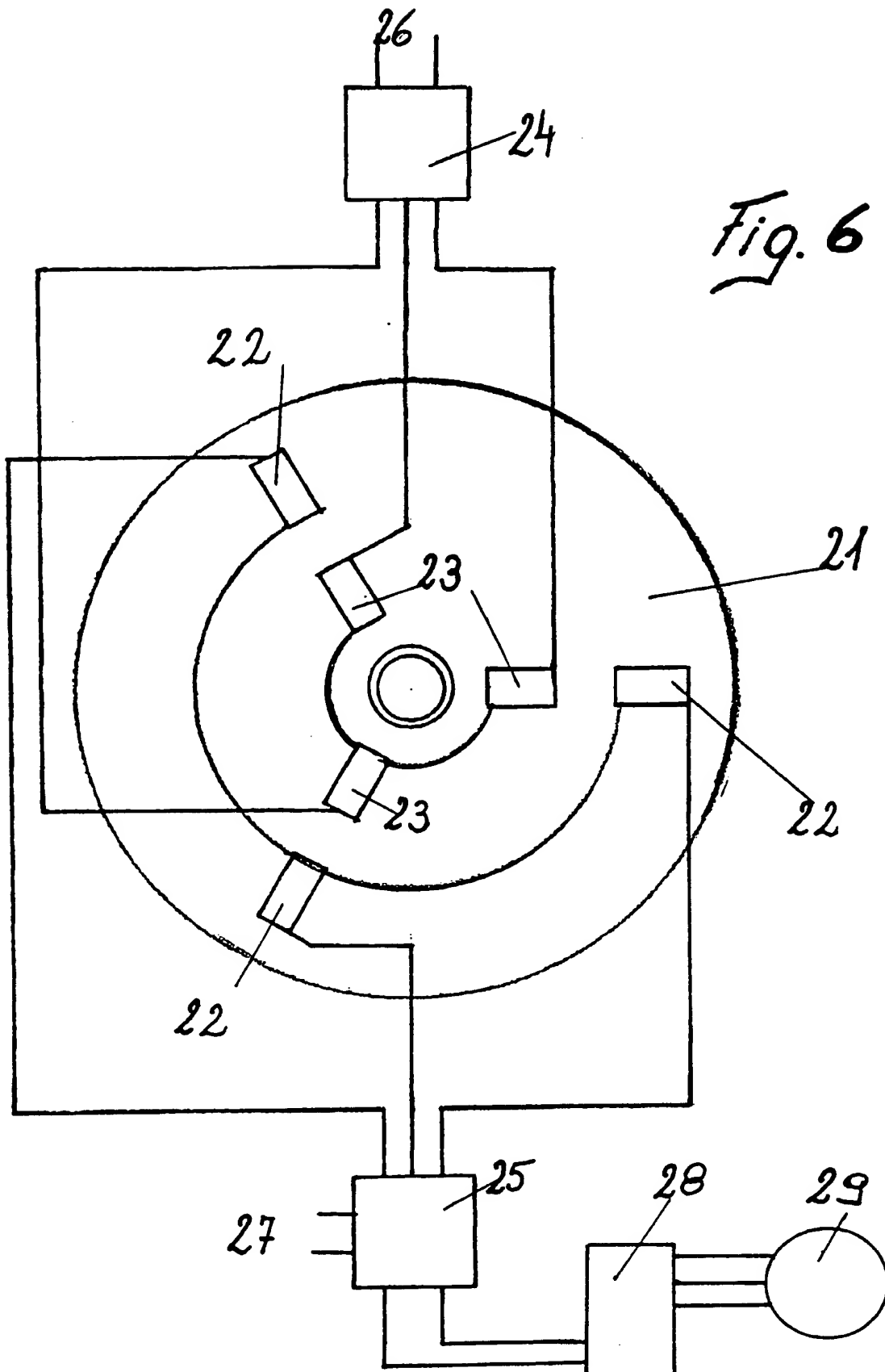
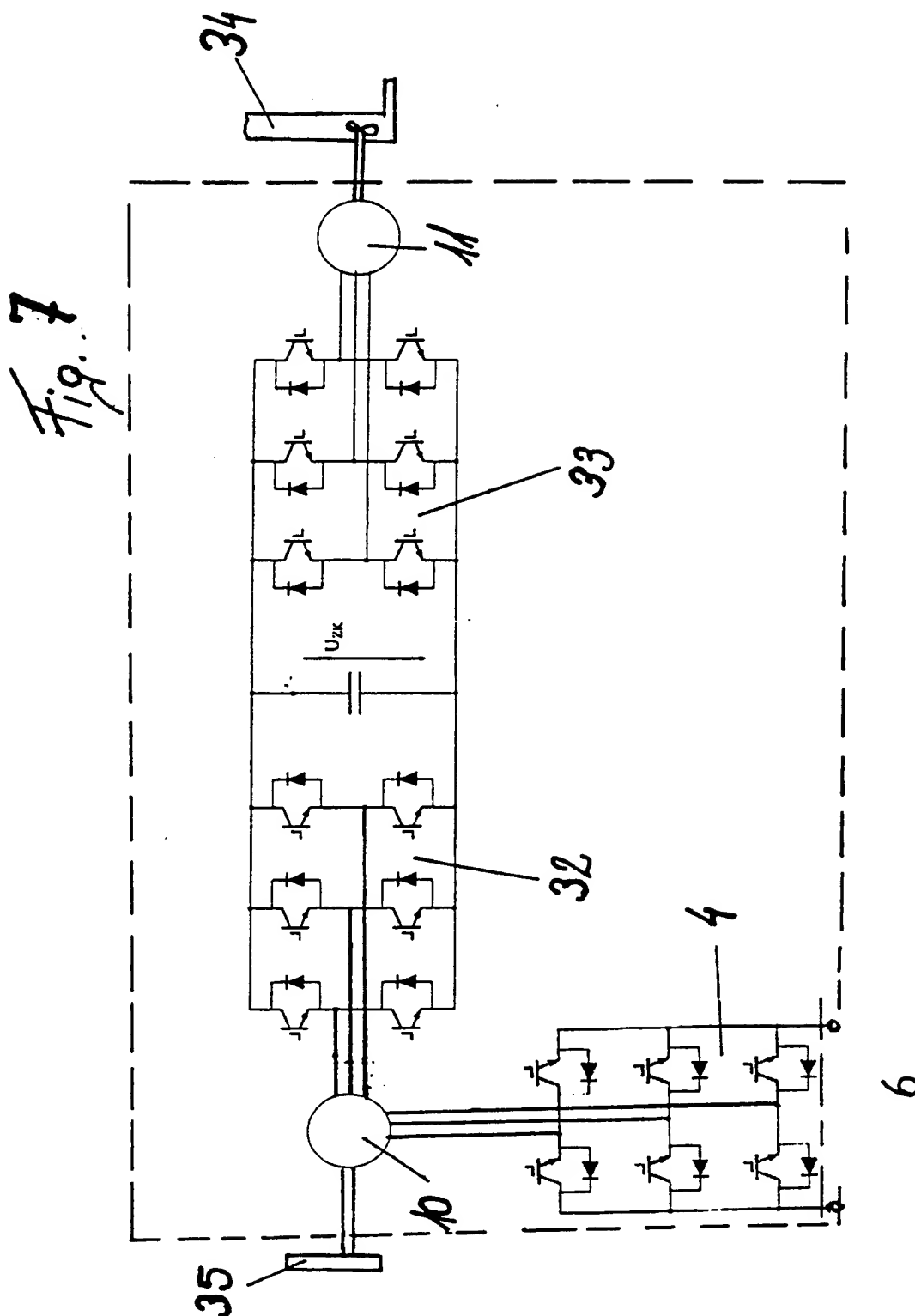


Fig. 5







INTERNATIONAL SEARCH REPORT

International Application No

PCT/AT 00/00167

A. CLASSIFICATION OF SUBJECT MATTER
 IPC 7 H02K16/00 H02K16/02

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
 IPC 7 H02K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 769 403 A (TOYOTA MOTOR CO LTD) 23 April 1997 (1997-04-23) paragraph '0006! column 8, line 1 - line 41; figures 1,2	1-3,6-8, 11
A	EP 0 725 474 A (NIPPON DENSO CO) 7 August 1996 (1996-08-07) column 4, line 12 - line 20 column 4, line 44 - line 51 column 5, line 15 - line 32 column 6, line 30 - line 33; figure 1	1-3,6-9
A	EP 0 800 951 A (TOYOTA MOTOR CO LTD) 15 October 1997 (15-10-1997) Abstract, Claim 1 figure 1	1-3,6-8

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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G document member of the same patent family

Date of the actual completion of the international search

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INTERNATIONALER RECHERCHENBERICHT

Internatio: : Aktenzeichen

PCT/AT 00/00167

A. KLASSIFIZIERUNG DES ANMELDUNGSGEGENSTANDES
IPK 7 H02K16/00 H02K16/02

Nach der Internationalen Patentklassifikation (IPK) oder nach der nationalen Klassifikation und der IPK

B. RECHERCHIERTE GEBIETE

Recherchierter Mindestprüfstoff (Klassifikationssystem und Klassifikationssymbole)

IPK 7 H02K

Recherchierte aber nicht zum Mindestprüfstoff gehörende Veröffentlichungen, soweit diese unter die recherchierten Gebiete fallen

Während der internationalen Recherche konsultierte elektronische Datenbank (Name der Datenbank und evtl. verwendete Suchbegriffe)

EPO-Internal, WPI Data, PAJ

C. ALS WESENTLICH ANGESEHENE UNTERLAGEN

Kategorie*	Bezeichnung der Veröffentlichung, soweit erforderlich unter Angabe der in Betracht kommenden Teile	Betr. Anspruch Nr.
A	EP 0 769 403 A (TOYOTA MOTOR CO LTD) 23. April 1997 (1997-04-23) Absatz '0006! Spalte 8, Zeile 1 - Zeile 41; Abbildungen 1,2	1-3,6-8, 11
A	EP 0 725 474 A (NIPPON DENSO CO) 7. August 1996 (1996-08-07) Spalte 4, Zeile 12 - Zeile 20 Spalte 4, Zeile 44 - Zeile 51 Spalte 5, Zeile 15 - Zeile 32 Spalte 6, Zeile 30 - Zeile 33; Abbildung 1	1-3,6-9
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☐ Weitere Veröffentlichungen sind der Fortsetzung von Feld C zu entnehmen

☒ Siehe Anhang Patentfamilie

* Besondere Kategorien von angegebenen Veröffentlichungen :

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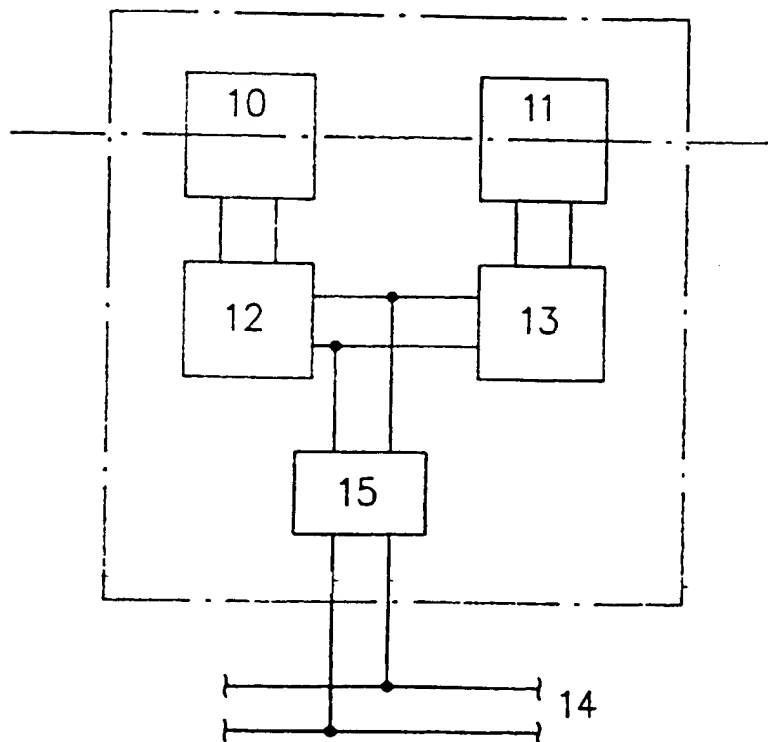
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(72) Erfinder: SCHRÖDL, Manfred [AT/AT]; Untere Haupt-
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(54) Title: ELECTRIC MOTOR

(54) Bezeichnung: ELEKTRISCHE MASCHINE



(57) Abstract: The invention relates to an electric motor, preferably of the three-phase current design. According to the invention, a first electric motor (10) is provided which is mechanically connected via the rotor (5) thereof to a rotating shaft of an engine, especially of an internal combustion engine. In addition, at least one second electric motor (11) is provided, whereby the second electric motor (11) is mechanically coupled via the rotor (6) thereof to a rotating part of a mechanical aggregate, especially to a turbo-engine. The first electric motor (10) is electrically coupled to at least the second electric motor (11) in order to exchange electrical power at a freely selectable voltage level.

(57) Zusammenfassung: Die Erfindung betrifft eine elektrische Maschine, vorzugsweise in Drehstromausführung. Es ist eine erste elektrische Maschine (10) vorgesehen, die über ihren Rotor (5) mit einer rotierenden Welle einer Maschine, insbesondere einer Verbrennungskraftmaschine, mechanisch verbunden ist. Ferner ist mindestens eine zweite elektrische Maschine (11) vorgesehen, wobei

die zweite elektrische Maschine (11) mit ihrem Rotor (6) mit einem rotierenden Teil eines mechanischen

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DK, DK (Gebrauchsmuster), DM, DZ, EE, EE (Gebrauchsmuster), ES, FI, FI (Gebrauchsmuster), GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SK (Gebrauchsmuster), SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.

(84) **Bestimmungsstaaten (regional):** ARIPO-Patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), eurasisches Patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), europäisches Patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI-Patent

(BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

Veröffentlicht:

- Mit internationalem Recherchenbericht.
- Vor Ablauf der für Änderungen der Ansprüche geltenden Frist; Veröffentlichung wird wiederholt, falls Änderungen eintreffen.

Zur Erklärung der Zweibuchstaben-Codes, und der anderen Abkürzungen wird auf die Erklärungen ("Guidance Notes on Codes and Abbreviations") am Anfang jeder regulären Ausgabe der PCT-Gazette verwiesen.

Aggregates, insbesondere einer Strömungsmaschine, mechanisch gekuppelt ist. Die erste elektrische Maschine (10) ist mit mindestens der zweiten elektrischen Maschine (11), zum Austausch elektrischer Energie auf frei wählbarem Spannungsniveau, elektrisch verbunden.

Elektrische Maschine

Die Erfindung betrifft eine elektrische Maschine, vorzugsweise in Drehstromausführung.

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Immer häufiger werden elektrische Maschinen in der Kraftfahrzeugtechnik eingesetzt. So sind Anordnungen, wie beispielsweise das ISAD-System (Integrated Starter-Alternator-Damper System) bekannt, das den Energieaustausch auf Bordspannungsniveau abwickelt.

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Ferner sind auch elektrisch betriebene Turbolader bekannt, bei denen ebenfalls der Energieaustausch auf Bordspannungsniveau durchgeführt wird. Dabei wird die Turbolader – Leistung zur Gänze aus dem Bordnetz entnommen.

15

Aufgabe der Erfindung ist es, eine elektrische Maschine zu schaffen, die insbesondere in der Kraftfahrzeugtechnik eingesetzt werden kann und die zur Versorgung zweier unterschiedlicher Netze, insbesondere für den Turbolader ausreichende elektrische Energie oder verschiedene Spannungsniveaus zur Verfügung stellt.

20

Die Aufgabe wird durch die Erfindung gelöst. Die erfindungsgemäße elektrische Maschine ist dadurch gekennzeichnet, daß eine erste elektrische Maschine vorgesehen ist, die über ihren Rotor mit einer rotierenden Welle einer Maschine, insbesondere einer Verbrennungskraftmaschine, mechanisch

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verbunden ist, daß mindestens eine zweite elektrische Maschine vorgesehen ist, daß die zweite elektrische Maschine mit ihrem Rotor mit einem rotierenden Teil eines mechanischen Aggregates, insbesondere einer Strömungsmaschine, mechanisch gekuppelt ist und daß die erste elektrische Maschine mit mindestens der zweiten elektrischen Maschine, zum Austausch elektrischer

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Energie auf frei wählbarem Spannungsniveau, elektrisch verbunden ist. Mit der Erfindung ist es erstmals möglich einen eigenen, autarken, internen elektrischen Kreis zu schaffen, der unabhängig vom Spannungsniveau des

Bordnetzes ist. Damit können die erfindungsgemäße Maschine und die leistungselektronischen Schaltungen mit optimalen Betriebsspannungen ausgelegt werden. Es ist ja bekannt, daß es üblicherweise vorteilhafter ist, mit höheren Spannungen als denen derzeit üblicher Bordnetze elektrische Leistung zu transportieren.

Dieser interne elektrische Teil ist mit der ersten Maschine über leistungselektronische Elemente wie Dioden und Transistoren gemäß dem Stand der Technik entsprechenden Schaltungen verbunden, um damit den internen elektrischen Teil in seinen elektrischen Kenngrößen, wie Spannungen und Ströme und deren zeitliche Verläufe zu gestalten.

Die erste Maschine kann mit dem Verbrennungsmotor mechanisches Drehmoment zu- und abführen, wodurch die erste Maschine bei Leistungsentnahme generatorisch arbeiten kann und diese Energie in Form elektrischer Energie an den internen elektrischen Teil abgibt. Wenn die erste Maschine vom internen elektrischen Teil Energie bezieht, arbeitet sie als Motor und kann dieses Drehmoment beispielsweise zum Starten der Verbrennungskraftmaschine oder zur Unterstützung oder Optimierung im Betrieb verwenden.

Nach einem besonderen Merkmal der Erfindung ist die erste elektrische Maschine mit ihrem Rotor mit einer Kurbelwelle oder einer mit der Kurbelwelle in mechanischer Verbindung stehenden Welle einer Verbrennungskraftmaschine mechanisch verbunden. Dadurch kann in einfachster Weise das mechanische Drehmoment zwischen der ersten elektrischen Maschine und der Verbrennungskraftmaschine ausgetauscht werden.

Gemäß einer Ausgestaltung der Erfindung ist die erste elektrische Maschine mit der Verbrennungskraftmaschine über ein Getriebe mechanisch verbunden.

Auch diese konstruktive Lösung verbessert bei einem elektrisch angetriebenen Turbolader das Drehmoment bei niedrigen Touren.

Nach einer weiteren Ausgestaltung der Erfindung ist die erste elektrische Maschine ein Teil der Verbrennungskraftmaschine, beispielsweise ist der Rotor in der ersten elektrischen Maschine in die Schwungscheibe der Verbrennungskraftmaschine integriert. Der Vorteil dieser konstruktiven Lösung liegt vor allem darin, daß die komplette Anordnung in einer relativ kleinen Bauweise hergestellt werden kann.

Nach einem besonderen Merkmal der Erfindung ist die erste elektrische Maschine mit mindestens einem externen elektrischen Kreis, vorzugsweise einem Bordnetz, verbunden. Diese zweite elektrische Koppelstelle ist über eine leistungselektronische Spannungsanpassschaltung entsprechend dem Stand der Technik mit dem Bordnetz verbunden. Damit kann Leistung zwischen dem internen elektrischen Teil und dem Bordnetz ausgetauscht werden. Damit kann die erste elektrische Maschine der erfindungsgemäßen Anordnung in der einen Energierichtung als Starter und in der anderen Energierichtung als Bordnetzladeeinrichtung betrieben werden.

Gemäß einer weiteren Ausgestaltung der Erfindung sind die erste und die zweite elektrische Maschine in einem Gehäuse angeordnet. Mit dieser Ausgestaltung ist es möglich, ein elektrisches Antriebssystem zu schaffen, das wirtschaftlich hergestellt und eingesetzt werden kann. Vorteilhaft bei dieser Erfindung ist, daß gegenüber bekannten elektromechanischen Antrieben oder rein mechanischen Antrieben, wie Getrieben, bei denen zwei verschiedene, vorzugsweise unabhängige Drehzahlen benötigt werden, wesentliche Teile, wie beispielsweise Gehäuseelemente, Teile der Steuerung eingespart werden können. Weiters können die bekannten EMV-Probleme lokal im Gehäuse gelöst werden und dringen nicht in die Umgebung.

Nach einer Weiterbildung der Erfindung ist bzw. sind die erste und/oder die zweite elektrische Maschine als Asynchron-, Synchron- oder Reluktanzmaschine ausgeführt. Dadurch kann für jeden Anwendungsfall die optimale Maschine gewählt werden.

- 5 Nach einem weiteren Merkmal der Erfindung weisen die erste und die zweite elektrische Maschine Rotoren mit gleicher Rotationsachse auf. Gerade in der Kraftfahrzeugtechnik ist es von Vorteil, wenn für eine mechanisch-elektrisch-mechanische Kupplung nur eine Rotationsachse gegeben ist.
- 10 Gemäß einer besonderen Ausgestaltung der Erfindung sind eine der beiden Maschinen als Innenläufer und die andere Maschine als Außenläufer ausgeführt. Auch durch diese Ausgestaltung der Erfindung ist eine kompakte Ausführung der Maschine möglich.
- 15 Nach einem weiteren Merkmal der Erfindung weisen die beiden elektrischen Maschinen ein gemeinsames Statorblechpaket auf. Bei dieser Ausgestaltung können in einem Gehäuse ein Stator mit mindestens einer Statorwicklung und mindestens zwei Rotoren vorgesehen sein. Die Rotoren sind mechanisch voneinander getrennt und jeder Rotor steht mit dem elektromagnetisch aktiven
- 20 Stator in elektromagnetischer Wechselwirkung, wobei die Drehzahlen der Rotoren gleich oder unterschiedlich sind.

- Gemäß einer Weiterbildung der Erfindung sind die Komponenten für den elektrischen Energieaustausch zwischen den elektrischen Maschinen und/oder
- 25 einem externen elektrischen Kreis in einem Gehäuse mindestens einer elektrischen Maschine angeordnet. Diese Weiterbildung trägt vor allem dazu bei, eine elektrische Maschine für die Kraftfahrzeugtechnik zu schaffen, die eine kompakte Bauweise aufweist.

- 30 Nach einer weiteren Ausgestaltung der Erfindung weist das Gehäuse mindestens einer elektrischen Maschine eine Flüssigkeitskühlung auf. Dadurch kann die Verlustwärme der Wicklungen, aber auch der leistungselektronischen

Elemente, die auf Grund der bekannten Probleme mit den hohen Strömen in der Maschine auftreten kann, optimal abgeführt werden.

5 Gemäß einem weiteren Merkmal der Erfindung ist vom elektrischen Kreis, der die beiden elektrischen Maschinen verbindet, ein Netzanschluß mit Gleich-, Wechsel- oder Drehspannung ableitbar. In einer dieser Ausgestaltung kann vom internen elektrischen Kreis ein weiteres Dreh-, Wechsel- oder Gleichspannungsnetz zur Verfügung gestellt werden. Beispielsweise kann ein kräftiges 230 V-Netz oder 3x400 V-Netz ausgekoppelt werden, wobei die
10 Frequenz entweder intern oder extern vorgegeben werden kann. Damit wird das Bordnetz und die an ihm angeschlossenen Aggregate über den internen elektrischen Kreis mit diesem Netz energiemäßig verbunden. Damit kann beispielsweise die Verbrennungskraftmaschine vom Netz gestartet werden, ohne das Bordnetz zu benötigen oder umgekehrt die
15 Verbrennungskraftmaschine ein bestehendes Netz stützen oder aufbauen. Es kann auch die Bordnetzatterie in einfacher Weise vom Netz geladen werden.

20 Nach einem besonderen Merkmal der Erfindung weist der Stator mindestens einer elektrischen Maschine mindestens zwei, vorzugsweise in der Maschine galvanisch getrennte, Wicklungssysteme auf, die mit dem Hauptfluß der Maschine magnetisch gekoppelt sind. Mit dieser Ausgestaltung der Erfindung ist es möglich, zwei autarke elektrische Kreise zu schaffen, die ein voneinander unabhängiges Spannungsniveau aufweisen. Ferner ist es ein Vorteil dieser Erfindung, daß elektro-magnetische, also EMV-Störungen durch das Schalten
25 in einem Wicklungssystem in einem anderen Wicklungssystem unterdrückt werden können. So können weiters auch vorteilhaft die einzelnen Wicklungssysteme auf verschiedenen Spannungsniveaus, insbesondere galvanisch trennbar, arbeiten. Eine eigene galvanische Trennung und/oder ein Transformator zur Spannungsanpassung zwischen den beiden beteiligten
30 Stromkreisen wird nicht mehr benötigt.

Gemäß einem besonderen Merkmal der Erfindung sind die mindestens zwei Wicklungssysteme über getrennte leistungselektronische Schaltungen mit jeweiligen, vorzugsweise galvanisch getrennten, Stromkreisen verbunden. Dadurch ist der Vorteil gegeben, daß beispielsweise ein Netz, insbesondere ein Bordnetz, von einem weiteren Netz getrennt betrieben und geregelt werden kann.

Nach einer weiteren Ausgestaltung der Erfindung ist mindestens ein Wicklungssystem über eine Gleichrichterbrücke mit einem Gleichspannungs- oder batteriegestützten Netz, vorzugsweise einem Bordnetz, zum Energieaustausch in einer Richtung verbunden ist. Durch diese Ausgestaltung können wirtschaftlichere, oder auch billigere, leistungselektronische Komponenten zum Laden verwendet werden.

Gemäß einem weiteren Merkmal der Erfindung ist mindestens ein Wicklungssystem über eine Transistorbrücke mit einem Gleichspannungs- oder batteriegestützten Netz, vorzugsweise einem Bordnetz, zum Energieaustausch in beiden Richtungen verbunden. Dadurch ist der Vorteil gegeben, daß auf einen eigenen Starter verzichtet werden kann oder es wird dem einen Netz, vorzugsweise dem Bordnetz Energie entnommen und das andere Netz gespeist.

Nach einem besonderen Merkmal der Erfindung ist mit mindestens einem der Wicklungssysteme die Maschine als Generator zum Laden des angeschlossenen Bordnetzes, sowie auch als Motor, vorzugsweise als Starter einer mechanisch gekoppelten Verbrennungskraftmaschine, betreibbar. Auch bei dieser Ausgestaltung ist der Vorteil gegeben, daß der Starter, aber auch die Lichtmaschine bei der Konzeption in Wegfall geraten können.

Gemäß einer Weiterbildung der Erfindung ist über die mindestens zwei Wicklungssysteme ein galvanisch trennbarer elektrischer Energieaustausch zwischen den an die Wicklungssysteme angeschlossenen Stromkreisen

durchführbar. Dadurch ist vorteilhafterweise eine Trennung des Bordnetzes vom zweiten Netz gegeben, das durchaus eine höhere Spannung aufweisen kann.

- 5 Nach einer weiteren Ausgestaltung der Erfindung übernehmen die über leistungselektronische steuerbare Schalter angesteuerten Wicklungssysteme die Führung der elektrischen Größen von über leistungselektronische, nicht steuerbare Elemente, vorzugsweise Dioden, gekoppelte Wicklungssysteme. Dabei ist vorteilhaft, daß zur Steuerung des Ladevorganges keine eigenen
- 10 steuerbaren Elemente notwendig sind, sondern auf die steuerbaren Elemente des zweiten Netzes zurückgegriffen werden kann.

- Gemäß einem weiteren Merkmal der Erfindung ist jedes Wicklungssystem galvanisch unabhängig vom jeweiligen anderen Wicklungssystem mit elektro-
- 15 mechanischen Funktionsgruppen auf im allgemeinen unterschiedlichen Spannungsebenen verbunden. Dadurch können die elektro-mechanischen Funktionsgruppen, wie eine elektrisch betriebene Ölpumpe oder Wasserpumpe, oder auch eine elektro-magnetisch betriebene Ventilsteuerung, für Ein- und Auslaßventile bzw. Motorventile, aber auch elektrisch betriebene Lüfter
- 20 unabhängig von der Leistungsbegrenzung der Gleichspannung bzw. der Batterie auf einem vorteilhaften Spannungs- und/oder Stromniveau betrieben werden.

- Nach einer besonderen Ausgestaltung der Erfindung ist durch enge
- 25 magnetische Koppelung von den Wicklungssystemen ein elektromagnetischer Energieaustausch zwischen diesen Wicklungssystemen unabhängig von einer Rotordrehung nach dem Prinzip des Transformators gegeben. Damit ist der Vorteil gegeben, daß auch bei stillstehenden Rotor über eine zeitvariable Spannung durch geeignete leistungselektronische Stellglieder an einem
- 30 Wicklungssystem ein Energietransport in das relativ eng gekoppelte andere Wicklungssystem möglich ist.

Gemäß einem weiteren Merkmal der Erfindung erfolgt durch schwache magnetische Koppelung von den Wicklungssystemen eine geringe elektromagnetische Beeinflussung der Wicklungssysteme. Dadurch ist der Vorteil gegeben, daß elektro-magnetische Störungen durch Schaltvorgänge in einem Wicklungssystem kaum im anderen Wicklungssystem wirksam werden.

Nach einer Weiterbildung der Erfindung ist durch Steuerung der elektromagnetischen Größen, vorzugsweise der Ströme und Flussverkettungen, mindestens eines Wicklungssystems ein beliebig gestaltbarer elektromechanischer Energieaustausch zwischen den Wicklungssystemen und der Rotorwelle erreichbar. Durch diese Ausgestaltung ist der Vorteil gegeben, daß mechanische und elektrische Energie entsprechend der aktuellen, optimalen Strategie zu Verfügung gestellt wird.

Gemäß einer weiteren Ausgestaltung der Erfindung sind eine erste und eine zweite elektrische Maschine in einem Gehäuse angeordnet. Mit dieser Ausgestaltung ist es möglich, ein elektrisches Antriebssystem zu schaffen, das wirtschaftlich hergestellt und eingesetzt werden kann. Vorteilhaft bei dieser Erfindung ist, daß gegenüber bekannten elektromechanischen Antrieben oder rein mechanischen Antrieben, wie Getrieben, bei denen zwei verschiedene, vorzugsweise unabhängige Drehzahlen benötigt werden, wesentliche Teile, wie beispielsweise Gehäuseelemente, Teile der Steuerung eingespart werden können. Weiters können die bekannten EMV-Probleme lokal im Gehäuse gelöst werden und dringen nicht in die Umgebung.

Nach einer Weiterbildung der Erfindung ist bzw. sind die erste und/oder die zweite elektrische Maschine als Asynchron-, Synchron- oder Reluktanzmaschine ausgeführt. Dadurch kann für jeden Anwendungsfall die optimale Maschine gewählt werden.

Nach einem weiteren Merkmal der Erfindung weisen die erste und die zweite elektrische Maschine Rotoren mit gleicher Rotationsachse auf. Gerade in der

Kraftfahrzeugtechnik ist es von Vorteil, wenn für eine mechanisch-elektrisch-mechanische Kupplung nur eine Rotationsachse gegeben ist.

Die Erfindung wird an Hand von Ausführungsbeispielen, die in der Zeichnung
5 dargestellt sind, näher erläutert.

Fig. 1 zeigt eine elektrische Maschine mit Rotoren mit gleicher Rotorachse,

Fig. 2 eine Prinzipskizze der elektrischen Schaltung der Maschine,
10

Fig. 3 die elektrische Maschine mit den elektronischen Elementen,

Fig. 4 und 5 eine Ausführungsvariante der elektrischen Maschine,

15 Fig. 6 eine Prinzipskizze einer elektrischen Schaltung der Maschine und

Fig. 7 eine Zusammenschaltung eines Generators mit einem Verdichtermotor
über Umrichter.

20 Einführend sei festgehalten, daß in der beschriebenen Ausführungsform gleiche
Teile bzw. Zustände mit gleichen Bezugszeichen bzw. gleichen
Bauteilbezeichnungen versehen werden, wobei die in der gesamten
Beschreibung enthaltenen Offenbarungen sinngemäß auf gleiche Teile bzw.
Zustände mit gleichen Bezugszeichen bzw. gleichen Bauteilbezeichnungen
25 übertragen werden können. Auch sind die in der Beschreibung gewählten
Lageangaben, wie z.B. oben, unten, seitlich usw. auf die unmittelbar
beschriebene sowie dargestellte Figur bezogen und sind bei einer
Lageänderung sinngemäß auf die neue Lage zu übertragen.

30 Weiters können auch Einzelmerkmale oder Merkmalskombinationen aus dem
gezeigten Ausführungsbeispielen für sich eigenständige, erfindungsgemäße

Lösungen darstellen. Die diesbezüglichen erfindungsgemäßen Aufgaben und Lösungen sind den detaillierten Beschreibungen dieser Figuren zu entnehmen.

Grundsätzlich sind verschiedene Varianten von derartigen elektrischen Maschinen möglich. Gemäß der Fig. 1 ist eine erste elektrische Maschine 10 mit einem Stator 1 der eine Wicklungen 2 aufweist, dargestellt. Bei der zylindrischen Motoranordnung befindet sich eine Wicklung 2 an der Innenseite des Stators 1 bzw. der Statorbohrung und kann als Nut- oder Luftspaltwicklung ausgeführt sein. Die zweite elektrische Maschine 11 weist eine Wicklung 3 an der Außenseite des Stators 4 als Nut- oder Luftspaltwicklung auf, wobei die Wicklung 2 mit einem als Innenläufer ausgeführten Rotor 5 sowie die Wicklung 3 mit einem als Außenläufer ausgebildeten Rotor 6 zusammenarbeitet. Die Rotoren 5, 6 können mit Permanentmagnetenerregung, als Käfigläufer, im Reluktanzaufbau, etc. ausgeführt sein. Die beiden Rotoren 5, 6 sind mechanisch über je eine geeignete Lagerung 7, 8 nach dem Stand der Technik im Gehäuse 9 gelagert.

Wie bereits erwähnt, soll das Haupteinsatzgebiet einer derartigen elektrischen Maschine 10, 11 in der Kraftfahrzeugtechnik liegen, wobei diese mehrere Funktionen erfüllen kann. So ist die erste elektrische Maschine 10 mit der Verbrennungskraftmaschine mechanisch gekuppelt, z.B. über ein Getriebe mit der Kurbelwelle oder die erste Maschine 10 befindet sich mit deren Rotor 5 direkt auf einem bestehenden Element des Verbrennungsmotors, wie etwa auf der Kupplungsschwungscheibe oder einem vorhandenen Abtriebsrad oder ist konstruktiv in diesen Teil integriert. Diese erste Maschine 10 kann mit dem Verbrennungsmotor daher mechanisches Drehmoment zu- und abführen, wodurch die erste Maschine 10 bei Leistungsentnahme generatorisch arbeiten kann und diese Energie in Form elektrischer Energie an den internen elektrischen Teil abgibt. Wenn die erste Maschine 10 vom internen elektrischen Teil Energie bezieht, arbeitet sie als Motor und kann dieses Drehmoment zum Starten der Verbrennungskraftmaschine oder zur Unterstützung oder Optimierung im Betrieb verwenden.

Gemäß der Fig. 2 ist die erste elektrische Maschine 10 sowie die zweite elektrische Maschine 11 jeweils mit einem Steuer- oder Leistungsteil 12, 13 verbunden. Zum Austausch elektrischer Energie auf frei wählbaren Spannungsniveau sind die beiden Steuer- und Leistungsteile 12, 13, die auch die elektronische Leistungsumformung durchführen miteinander verbunden. Dieser interne elektrische Kreis ist mit der ersten Maschine über leistungselektronische Elemente, wie Dioden und Transistoren in dem Stand der Technik entsprechenden Schaltungen verbunden, um damit den internen elektrischen Teil in seinen elektrischen Kenngrößen, wie Spannungen und Ströme und deren zeitliche Verläufe zu gestalten. Ein wichtiges Merkmal dieses internen elektrischen Kreises und damit des Spannungsniveaus der ersten Maschine ist die Unabhängigkeit des Spannungsniveaus von einem externen, elektrischen Kreis, dem sogenannten Bordnetz 14. Damit können die erfindungsgemäßen Maschinen und leistungselektronischen Schaltungen mit optimalen Betriebsspannungen ausgelegt werden. Es ist bekannt, daß es üblicherweise vorteilhafter ist, mit höheren Spannungen als denen derzeit üblicher Bordnetze elektrische Leistung zu transportieren.

Dieses externe Bordnetz 14 ist über einen weiteren Steuer- bzw. Leistungsteil 15 mit dem internen elektrischen Kreis verbunden.

Darin ist auch ein wesentlicher Vorteil dieser Anordnung gegeben, da der mechanischen Energieaustausch zwischen Verbrennungsmotor und einem oder mehreren weiteren Aggregaten wie Turboladern, Pumpen, Lüftern, Kompressoren etc. ohne Verwendung des Bordnetzes abgewickelt werden kann. Neben dem optimalen Spannungsniveau bietet die Anordnung auch ein wesentlich besseres EMV-Verhalten, da die EMV-Störungen durch einfache Maßnahmen entsprechend dem Stand der Technik nicht an das Bordnetz 14 bzw. allgemein in die Umgebung des Aggregats gelangen, sondern nur innerhalb des Aggregats bewältigt werden müssen. Weiters kann mit der Anordnung wesentlich mehr Energie zur Speisung von Hilfsaggregaten als über das Bordnetz drehzahlunabhängig übertragen werden.

Von diesem internen elektrischen Teil, dessen Spannung an den optimalen Betrieb der Anordnung laufend angepaßt werden kann, sofern die leistungselektronischen Elemente dies ermöglichen, gehen nun eine oder vorzugsweise zwei oder mehrere elektrische Leistungsaustausch-Koppelstellen aus.

Die erste elektrische Koppelstelle geht über leistungselektronische Elemente zum elektrischen Anschluß der zweiten Maschine 11, die elektrische Leistung auf von der ersten Maschine 10 grundsätzlich unabhängigem Drehzahlniveau, in mechanische Leistung umwandeln kann. Diese mechanische Leistung dient in einer bevorzugten Variante dieser Anordnung dazu, eine Strömungsmaschine, wie beispielsweise einen Turbolader zu betreiben, um damit den Vorteil eines verbrennungsmotordrehzahlunabhängigen Betriebes der Strömungsmaschine zu ermöglichen. Gegenüber bekannten elektrisch betriebenen Turboladern weist diese Anordnung ferner den großen Vorteil auf, daß die Turbolader-Leistung nicht notwendigerweise zur Gänze aus dem Bordnetz 14 entnommen wird, sondern ganz oder teilweise über die erste Maschine 10 mit dem Verbrennungsmotor ausgetauscht wird. Dies entlastet das Bordnetz massiv und erlaubt Energieaustausch auf günstigerem elektrischen Spannungsniveau, wodurch die Verkabelung und die leistungselektronischen Komponenten vorteilhafter ausgeführt werden können. Der Leistungsaustausch ist bidirektional möglich. In gleicher Art können weitere elektrische Maschinen als Bestandteil der Erfindung zum Antrieb weiterer Aggregate wie Wasserpumpen, Lüfter, Kompressoren usw. am internen elektrischen Teil angekoppelt sein.

Die zweite elektrische Koppelstelle ist über eine leistungselektronische Spannungsanpaßschaltung entsprechend dem Stand der Technik mit dem Bordnetz verbunden. Damit kann Leistung zwischen dem internen elektrischen Teil und dem Bordnetz 14 ausgetauscht werden. Damit kann die erste elektrische Maschine 10 der Anordnung in der einen Energierichtung als Starter und in der anderen Energierichtung als Bordnetzladeeinrichtung betrieben

werden. Der große Vorteil dieses Betriebsmodus ist, daß kein eigener Starter und keine eigene Lichtmaschine nötig sind, weil diese Funktionen von der Anordnung abgedeckt werden. Ein wesentlicher Vorteil gegenüber bekannten Anordnungen ist, daß der Starter nun als Maschine mit optimalem

5 Spannungsniveau ausgelegt und betrieben werden kann, so daß die bekannten Probleme mit hohen Strömen in der Maschine und auch in der an den Maschinensträngen angeschlossenen leistungselektronischen Elementen vermieden werden.

10 Weiters kann die zweite Maschine 11 über das Bordnetz 14 unabhängig vom Verbrennungsmotor, also etwa auch während dessen Stillstand, betrieben werden. Damit kann beispielsweise der Turbolader im Stillstand der Verbrennungskraftmaschine gestartet werden, um einen besseren Startprozess zu ermöglichen. Gegenüber bekannten Lösungen bietet diese Lösung den

15 Vorteil, daß der zweite Motor und die motornahe Leistungselektronik für ein optimales Spannungsniveau ausgelegt und betrieben werden kann.

In einer weiteren Ausgestaltung kann vom internen elektrischen Kreis ein weiteres Dreh-, Wechsel- oder Gleichspannungsnetz zur Verfügung gestellt
20 werden. Beispielsweise kann ein kräftiges 230 V-Netz oder 3x400 V-Netz ausgekoppelt werden, wobei die Frequenz entweder intern oder extern vorgegeben werden kann. Damit wird das Bordnetz 14 und die an ihm angeschlossenen Aggregate über den internen elektrischen Kreis mit diesem Netz energiemäßig verbunden.

25 Damit kann beispielsweise die Verbrennungskraftmaschine vom Netz gestartet werden, ohne das Bordnetz zu benötigen oder umgekehrt die Verbrennungskraftmaschine ein bestehendes Netz stützen oder aufbauen. Es kann auch die Bordnetzatterie in einfacher Weise vom Netz geladen werden.

30 Entsprechend der Fig. 3 ist die erste elektrische Maschine 10 und die zweite elektrische Maschine 11 im Gehäuse 9 angeordnet.

Zur Kühlung kann das Gehäuse 9 Kühlkanäle 16 aufweisen.

5 Zweckmäßigerweise kann im Bereich dieser gut gekühlten Gehäuseteile eine leistungs- und steuerungelektronische Schaltung einschließlich elektrischer, magnetischer und mechanischer Bauteile, wie Halbleiter 19, Kondensatoren 18, Drosseln, Relais od. dgl. sowie allfälliger Trägermaterialien 17 angeordnet sein, um die Funktionen gemäß den Elementen, wie dem Steuer- und Leistungsteil 12, 13, 15 in Fig. 2, vorteilhaft zu realisieren.

10 Eine weitere Variante der Bauweise der elektrischen Maschine ist aus der Fig. 4 und der Fig. 5 zu entnehmen. Dabei können die beiden elektrischen Maschinen 10, 11 übereinander angeordnet sein und der Rotorausgang kann bei jeder Maschine links und/oder rechts vorgesehen werden. Dabei kann in diesem Maschinengehäuse auch der elektronische Teil 20 integriert sein.

15 Gemäß der Fig. 6 ist eine elektrische Maschine in Drehstromausführung dargestellt, wobei diese elektrische Maschine die erste oder die zweite elektrische Maschine sein kann. Der Rotor der ersten elektrischen Maschine ist beispielsweise mit einer rotierenden Welle einer Verbrennungskraftmaschine mechanisch verbunden. Der Rotor der zweiten elektrischen Maschine ist mit
20 einem rotierenden Teil, beispielsweise einer Strömungsmaschine, gekuppelt. Zum Austausch elektrischer Energie auf frei wählbaren Spannungsniveau ist die erste elektrische Maschine mit der zweiten elektrischen Maschine elektrisch verbunden.

25 Der Stator 21 mindestens einer der beiden elektrischen Maschinen weist mindestens zwei Wicklungssysteme 22 bzw. 23 auf. Die beiden Wicklungssysteme 22, 23 sind in der elektrischen Maschine vorzugsweise galvanisch getrennt und sind mit dem Hauptfluß der Maschine magnetisch gekoppelt. Durch die galvanische Trennung, das heißt jedes Wicklungssystem
30 22, 23 liegt vorzugsweise in seinen eigenen Nuten, können EMV-Störungen durch das Schalten in einem Wicklungssystem 22, 23 unterdrückt werden.

Die beiden Wicklungssysteme 22, 23 sind über getrennte leistungselektronische Schaltungen 24, 25 mit jeweiligen, ebenfalls vorzugsweise galvanisch getrennten, Stromkreisen verbunden. So kann das Wicklungssystem 22 über die leistungselektronische Schaltung 24, beispielsweise einer

5 Gleichrichterbrücke oder mit einer Transistorbrücke mit einem Gleichspannungs- oder batteriegestützten Netz, vorzugsweise mit dem Bordnetz 26, zum Energieaustausch in einer oder beiden Richtungen verbunden sein. Natürlich könnte dieses Wicklungssystem 22 auch als Motor, vorzugsweise als Starter einer Verbrennungskraftmaschine, betreibbar sein.

10 Über die leistungselektronische Schaltung 25 kann ein Netz 27 gespeist werden. Ebenso ist aber auch diese leistungselektronische Schaltung 25 über das interne Netz mit einem leistungselektronischen Stellglied 28 für die zweite elektrische Maschine 29 elektrisch verbunden sein.

15 Jedes Wicklungssystem 22, 23 ist galvanisch unabhängig vom jeweiligen anderen Wicklungssystem 22, 23 mit elektro-mechanischen Funktionsgruppen auf im allgemeinen unterschiedlichen Spannungsebenen verbunden. Dadurch können die elektro-mechanischen Funktionsgruppen, wie eine elektrisch

20 betriebene Ölpumpe oder Wasserpumpe, oder auch eine elektro-magnetisch betriebene Ventilsteuerung, für Ein- und Auslaßventile bzw. Motorventile, aber auch elektrisch betriebene Lüfter unabhängig von der Leistungsbegrenzung der Gleichspannung bzw. der Batterie auf einem vorteilhaften Spannungs- und/oder Stromniveau betrieben werden.

25 Die Wicklungssysteme 22, 23 können eine schwache magnetische Kopplung aufweisen, beispielsweise wenn die Wicklungssysteme in verschiedenen Nuten untergebracht sind, oder auch eine enge magnetische Kopplung aufweisen, wenn beide Wicklungssysteme 22, 23 in einer Nut angeordnet werden.

30 Gemäß der Fig. 7 ist ein Generator, beispielsweise als erste elektrische Maschine 10 und ein Verdichtermotor als zweite elektrische Maschine 11

aufgezeigt. Die beiden elektrischen Maschinen sind über einen Generator-Umrichter 32 und einem Verdichtermotor-Umrichter 33 elektrisch miteinander verbunden. Mit Uzk ist dabei die Zwischenkreisspannung bezeichnet.

- 5 Der Generator ist mit seinem Rotor mit einer Maschine, insbesondere einer Brennkraftmaschine über ein Getriebe 35 verbunden. Der Verdichtermotor 31 ist mit seinem Rotor mit einer Strömungsmaschine 34 verbunden. Ein Wicklungssystem 22, 23 ist über eine leistungselektronische Schaltung 4 mit einem Bordnetz 6 verbunden, wobei die Wicklungssysteme 22, 23 galvanisch
10 trennbar sind.

Dabei könnte die erste und die zweite elektrische Maschine in einem Gehäuse angeordnet sein. Ebenfalls könnte die erste und die zweite elektrische Maschine Rotoren mit gleicher Rotationsachse aufweisen.

- 15 Abschließend sei der Ordnung halber darauf hingewiesen, daß in der Zeichnung einzelne Bauteile und Baugruppen zum besseren Verständnis der Erfindung unpropotional und maßstäblich verzerrt dargestellt sind.

20

25

Patentansprüche:

1. Elektrische Maschine, vorzugsweise in Drehstromausführung, dadurch gekennzeichnet, daß eine erste elektrische Maschine (10) vorgesehen ist,
5 die über ihren Rotor (5) mit einer rotierenden Welle einer Maschine, insbesondere einer Verbrennungskraftmaschine, mechanisch verbunden ist, daß mindestens eine zweite elektrische Maschine (11) vorgesehen ist, daß die zweite elektrische Maschine (11) mit ihrem Rotor (6) mit einem rotierenden Teil eines mechanischen Aggregates, insbesondere einer
10 Strömungsmaschine, mechanisch gekuppelt ist und daß die erste elektrische Maschine (10) mit mindestens der zweiten elektrischen Maschine (11), zum Austausch elektrischer Energie auf frei wählbarem Spannungsniveau, elektrisch verbunden ist.
- 15 2. Elektrische Maschine nach Anspruch 1, dadurch gekennzeichnet, daß die erste elektrische Maschine (10) mit ihrem Rotor (5) mit einer Kurbelwelle oder einer mit der Kurbelwelle in mechanischer Verbindung stehenden Welle einer Verbrennungskraftmaschine mechanisch verbunden ist.
- 20 3. Elektrische Maschine nach Anspruch 1, dadurch gekennzeichnet, daß die erste elektrische Maschine (10) mit der Verbrennungskraftmaschine über ein Getriebe mechanisch verbunden ist.
- 25 4. Elektrische Maschine nach Anspruch 1, dadurch gekennzeichnet, daß die erste elektrische Maschine (10) ein Teil der Verbrennungskraftmaschine ist, beispielsweise daß der Rotor (5) der ersten elektrischen Maschine (10) in die Schwungscheibe der Verbrennungskraftmaschine integriert ist.
- 30 5. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 4, dadurch gekennzeichnet, daß die erste elektrische Maschine (10) mit mindestens einem externen elektrischen Kreis, vorzugsweise einem Bordnetz (14), verbunden ist.

6. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 5, dadurch gekennzeichnet, daß die erste (10) und die zweite elektrische Maschine (11) in einem Gehäuse (9) angeordnet sind.
- 5 7. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 6, dadurch gekennzeichnet, daß die erste (10) und/oder die zweite elektrische Maschine (11) als Asynchron-, Synchron- oder Reluktanzmaschine ausgeführt ist bzw. sind.
- 10 8. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 7, dadurch gekennzeichnet, daß die erste (10) und die zweite elektrische Maschine (11) Rotoren (5, 6) mit gleicher Rotationsachse aufweisen.
- 15 9. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 8, dadurch gekennzeichnet, daß eine der beiden Maschinen (10, 11) als Innenläufer und die andere Maschine als Außenläufer ausgeführt sind.
- 20 10. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 9, dadurch gekennzeichnet, daß die beiden elektrischen Maschinen (10, 11) ein gemeinsames Statorblechpaket aufweisen.
- 25 11. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 10, dadurch gekennzeichnet, daß die Komponenten für den elektrischen Energieaustausch zwischen den elektrischen Maschinen (10, 11) und/oder einem externen elektrischen Kreis (14) in einem Gehäuse (9) mindestens einer elektrischen Maschine (10, 11) angeordnet sind.
- 30 12. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 11, dadurch gekennzeichnet, daß das Gehäuse (9) mindestens einer elektrischen Maschine (10, 11) eine Flüssigkeitskühlung aufweist.

13. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 12, dadurch gekennzeichnet, daß vom elektrischen Kreis, der die beiden elektrischen Maschinen verbindet, ein Netzanschluß mit Gleich-, Wechsel- oder Drehspannung ableitbar ist.

5

14. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 13, dadurch gekennzeichnet, daß der Stator (1, 4) mindestens einer elektrischen Maschine (10, 11) mindestens zwei, vorzugsweise in der Maschine (10, 11) galvanisch getrennte, Wicklungssysteme (22, 23) aufweist, die mit dem Hauptfluß der Maschine (10, 11) magnetisch gekoppelt sind.

10

15. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 14, dadurch gekennzeichnet, daß die mindestens zwei Wicklungssysteme (22, 23) über getrennte leistungselektronische Schaltungen (24, 25) mit jeweiligen, vorzugsweise galvanisch getrennten, Stromkreisen verbunden sind.

15

16. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 15, dadurch gekennzeichnet, daß mindestens ein Wicklungssystem (22, 23) über eine Gleichrichterbrücke mit einem Gleichspannungs- oder batteriegestützten Netz, vorzugsweise einem Bordnetz (26), zum Energieaustausch in einer Richtung verbunden ist.

20

17. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 16, dadurch gekennzeichnet, daß mindestens ein Wicklungssystem (22, 23) über eine Transistorbrücke mit einem Gleichspannungs- oder batteriegestützten Netz, vorzugsweise einem Bordnetz (26), zum Energieaustausch in beiden Richtungen verbunden ist.

25

30

18. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 17, dadurch gekennzeichnet, daß mit mindestens einem der Wicklungssysteme

(22, 23) die Maschine als Generator zum Laden des angeschlossenen Bordnetzes (26), sowie auch als Motor, vorzugsweise als Starter einer mechanisch gekoppelten Verbrennungskraftmaschine, betreibbar ist.

- 5 19. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 18, dadurch gekennzeichnet, daß über die mindestens zwei Wicklungssysteme (22, 23) ein galvanisch trennbarer elektrischer Energieaustausch zwischen den an die Wicklungssysteme (22, 23) angeschlossenen Stromkreisen durchführbar ist.
- 10 20. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 19, dadurch gekennzeichnet, daß die über leistungselektronische steuerbare Schalter angesteuerten Wicklungssysteme (22, 23) die Führung der elektrischen Größen von über leistungselektronische, nicht steuerbare
- 15 Elemente, vorzugsweise Dioden, gekoppelte Wicklungssysteme (22, 23) übernehmen.
21. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 20, dadurch gekennzeichnet, daß jedes Wicklungssystem (22, 23) galvanisch
- 20 unabhängig vom jeweiligen anderen Wicklungssystem (22, 23) mit elektromechanischen Funktionsgruppen auf im allgemeinen unterschiedlichen Spannungsebenen verbunden ist.
22. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 21,
- 25 dadurch gekennzeichnet, daß durch enge magnetische Koppelung von den Wicklungssystemen (22, 23) ein elektromagnetischer Energieaustausch zwischen diesen Wicklungssystemen (22, 23) unabhängig von einer Rotordrehung nach dem Prinzip des Transformators gegeben ist.
- 30 23. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 22, dadurch gekennzeichnet, daß durch schwache magnetische Koppelung von

den Wicklungssystemen (22, 23) eine geringe elektromagnetische Beeinflussung der Wicklungssysteme (22, 23) erfolgt.

24. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 23,
5 dadurch gekennzeichnet, daß durch Steuerung der elektromagnetischen Größen, vorzugsweise der Ströme und Flussverkettungen, mindestens eines Wicklungssystems (22, 23) ein beliebig gestaltbarer elektromechanischer Energieaustausch zwischen den Wicklungssystemen (22, 23) und der Rotorwelle erreichbar ist.
- 10 25. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 24, dadurch gekennzeichnet, daß eine erste und die zweite elektrische Maschine (10, 11) in einem Gehäuse angeordnet sind.
- 15 26. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 25, dadurch gekennzeichnet, daß die erste und/oder die zweite elektrische Maschine (10, 11) als Asynchron-, Synchron- oder Reluktanzmaschine ausgeführt ist bzw. sind.
- 20 27. Elektrische Maschine nach mindestens einem der Ansprüche 1 bis 26, dadurch gekennzeichnet, daß die erste und die zweite elektrische Maschine (10, 11) Rotoren mit gleicher Rotationsachse aufweisen.

Fig. 1

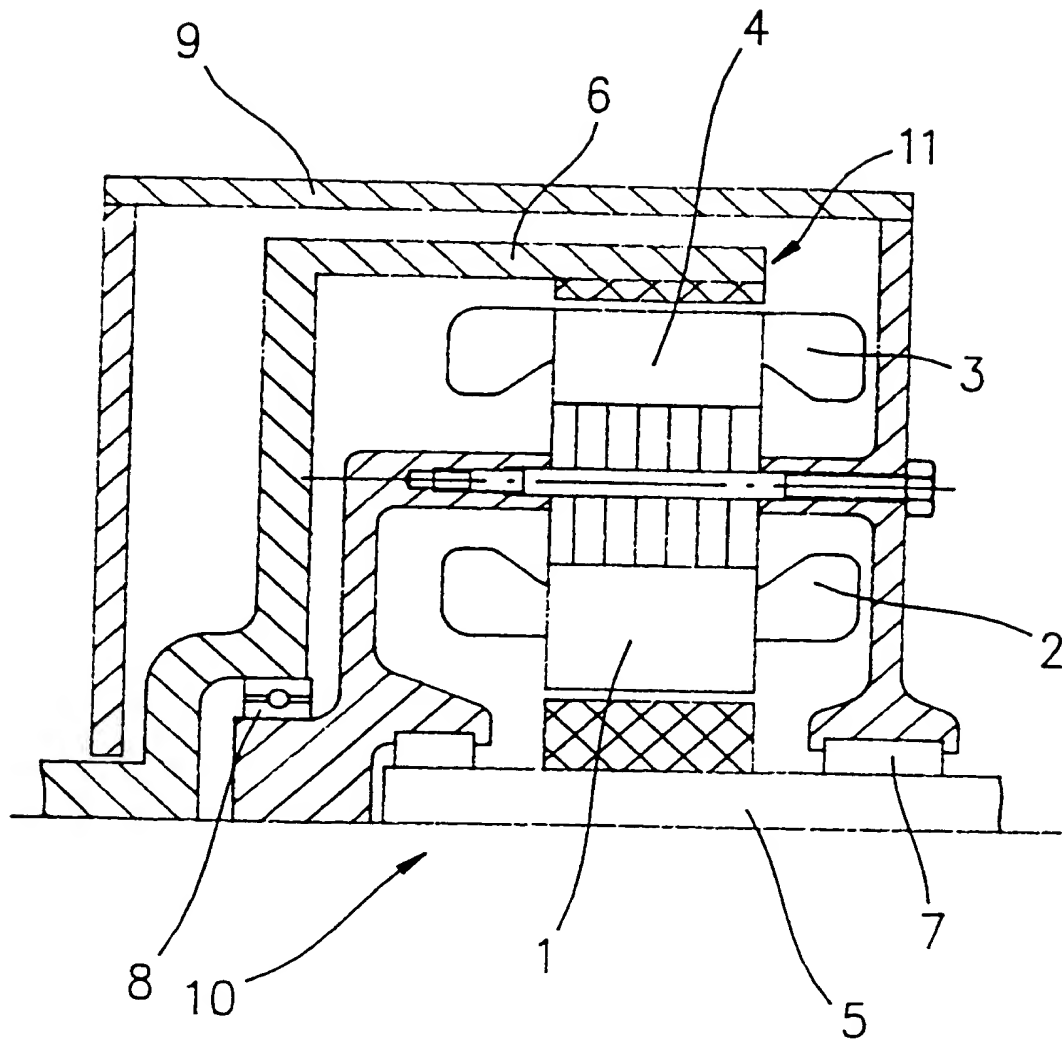


Fig. 2

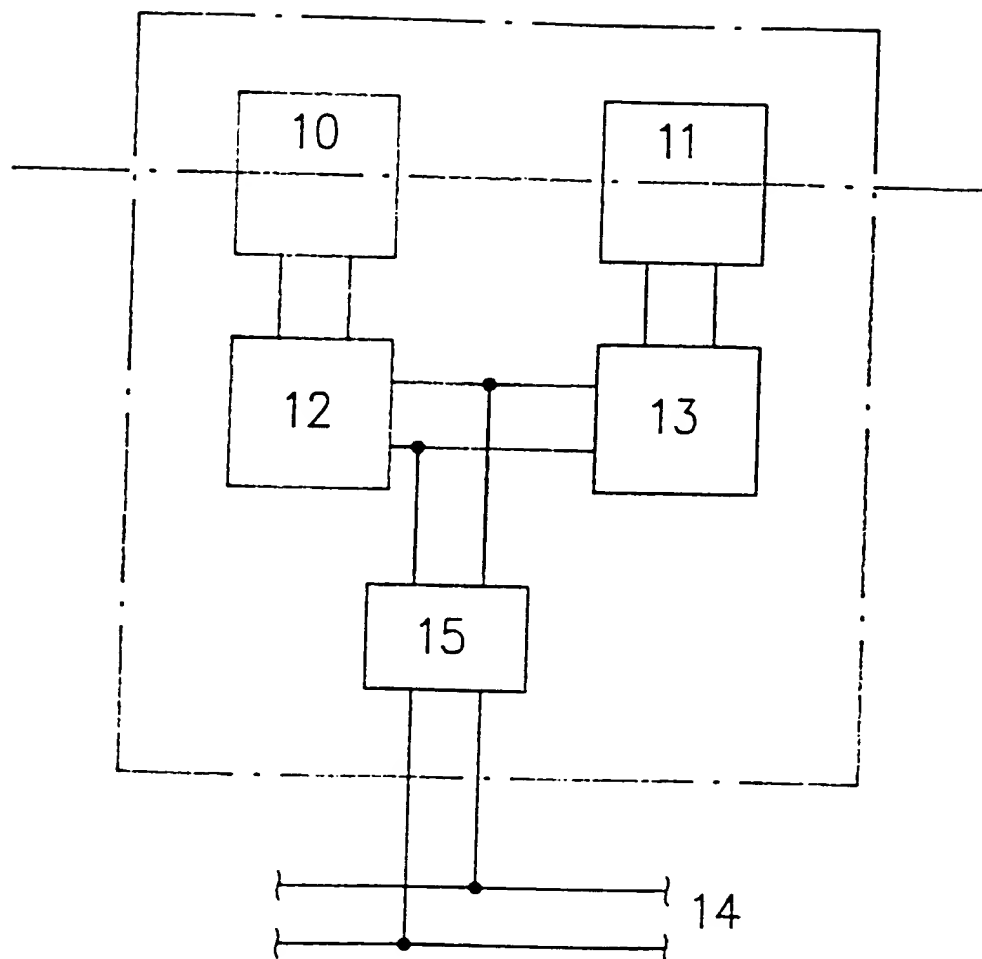


Fig. 3

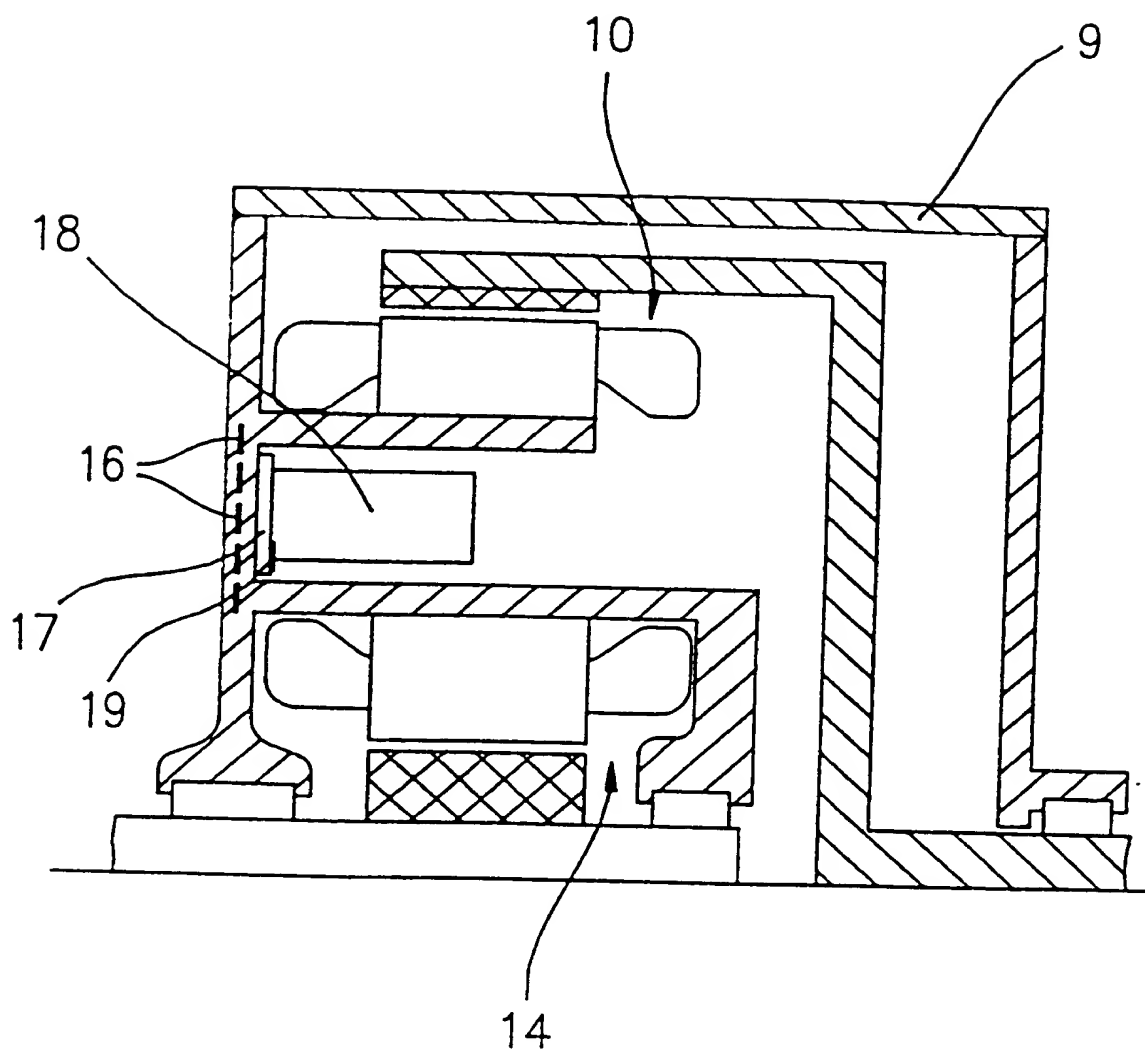


Fig. 4

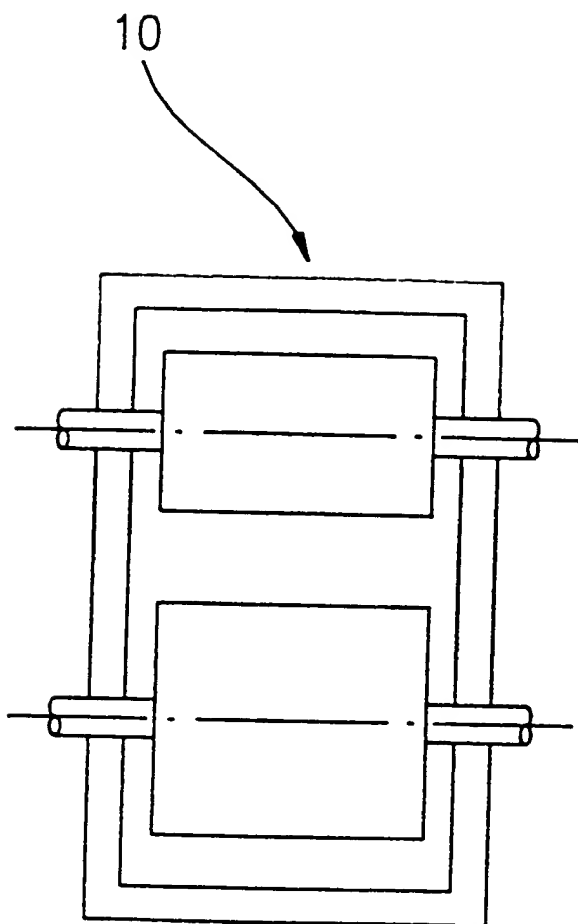
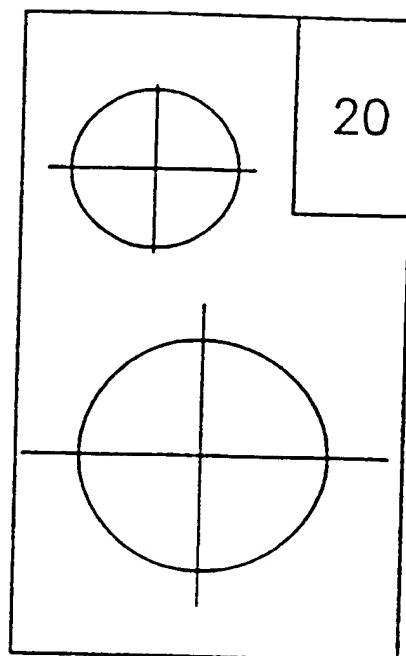


Fig. 5



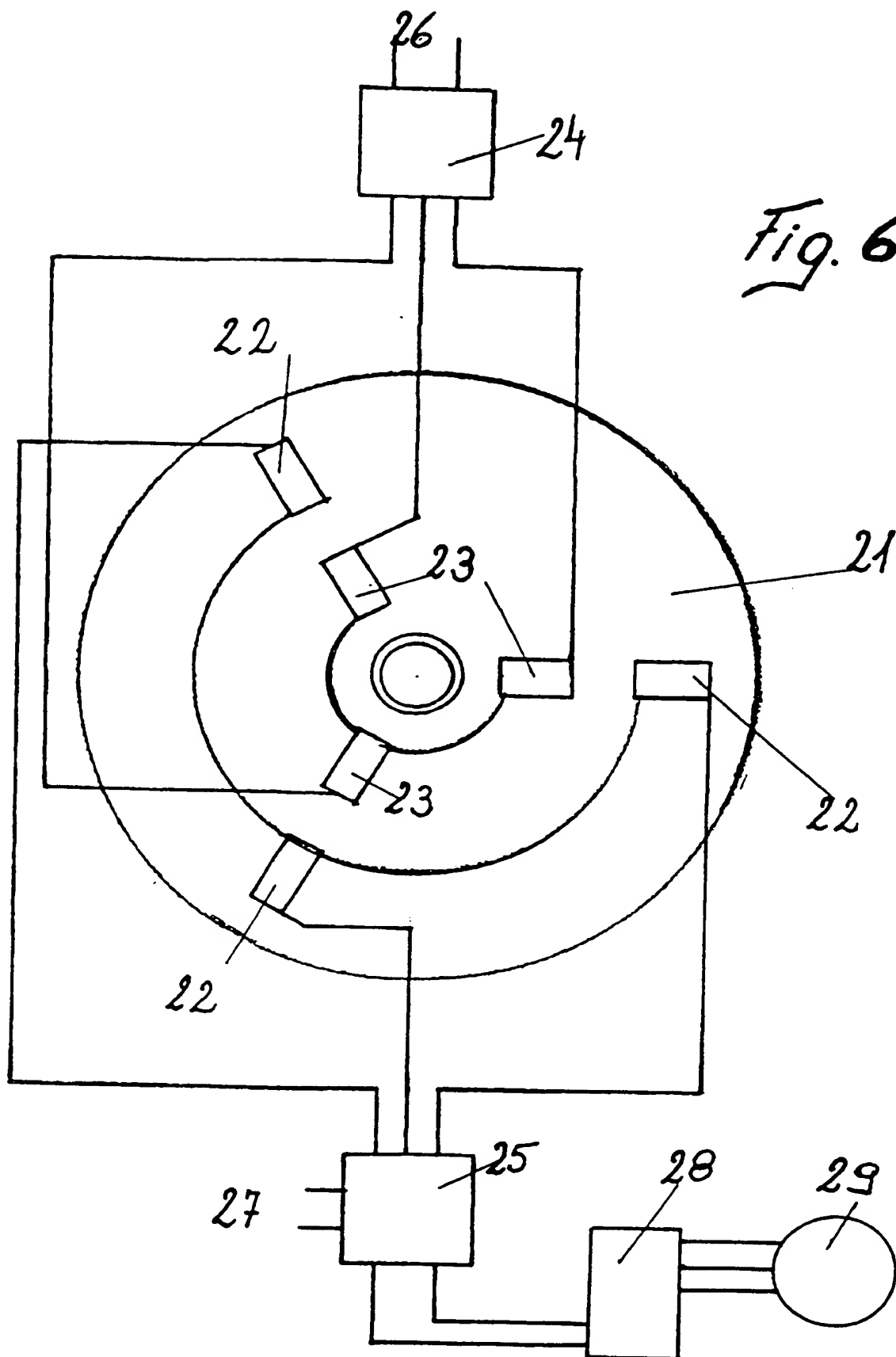
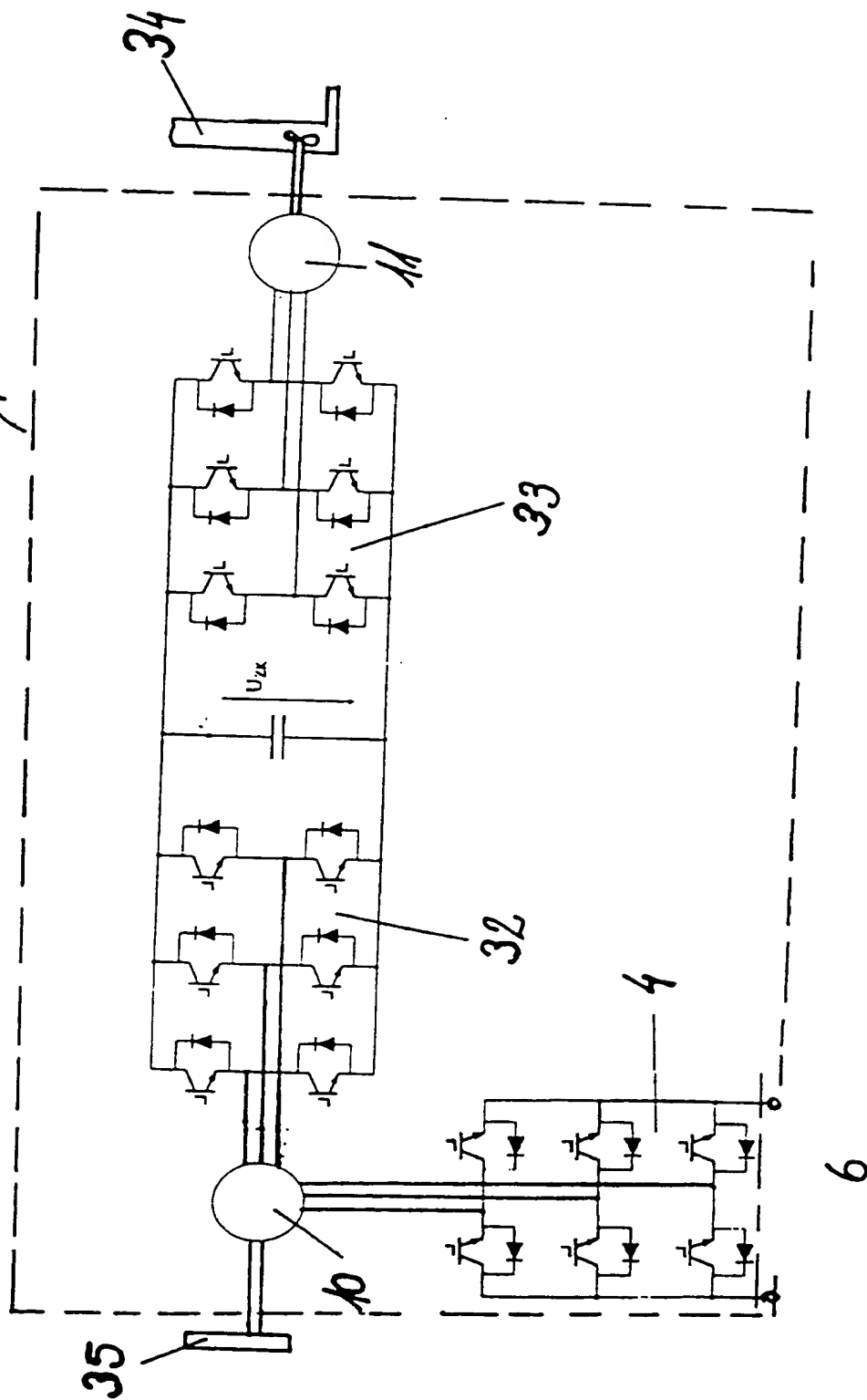


Fig. 7



INTERNATIONAL SEARCH REPORT

International Application No.

PCT/AT 00/00167

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 H02K16/00 H02K16/02

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHEDMinimum documentation searched (classification system followed by classification symbols)
IPC 7 H02K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)
EPO-Internal, WPI Data, PAJ**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 769 403 A (TOYOTA MOTOR CO LTD) 23 April 1997 (1997-04-23) paragraph '0006! column 8, line 1 - line 41; figures 1,2	1-3,6-8, 11
A	EP 0 725 474 A (NIPPON DENSO CO) 7 August 1996 (1996-08-07) column 4, line 12 - line 20 column 4, line 44 - line 51 column 5, line 15 - line 32 column 6, line 30 - line 33; figure 1	1-3,6-9
A	EP 0 800 951 A (TOYOTA MOTOR CO LTD) 15 October 1997 (15-10-1997) Abstract, Claim 1 figure 1	1-3,6-8

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C. ALS WESENTLICH ANGESEHENE UNTERLAGEN

Kategorie*	Bezeichnung der Veröffentlichung, soweit erforderlich unter Angabe der in Betracht kommenden Teile	Betr. Anspruch Nr.
A	EP 0 769 403 A (TOYOTA MOTOR CO LTD) 23. April 1997 (1997-04-23) Absatz '0006! Spalte 8, Zeile 1 - Zeile 41; Abbildungen 1,2	1-3,6-8, 11
A	EP 0 725 474 A (NIPPON DENSO CO) 7. August 1996 (1996-08-07) Spalte 4, Zeile 12 - Zeile 20 Spalte 4, Zeile 44 - Zeile 51 Spalte 5, Zeile 15 - Zeile 32 Spalte 6, Zeile 30 - Zeile 33; Abbildung 1	1-3,6-9
A	EP 0 800 951 A (TOYOTA MOTOR CO LTD) 15. Oktober 1997 (1997-10-15) Zusammenfassung, Anspruch 1 Abbildung 1	1-3,6-8

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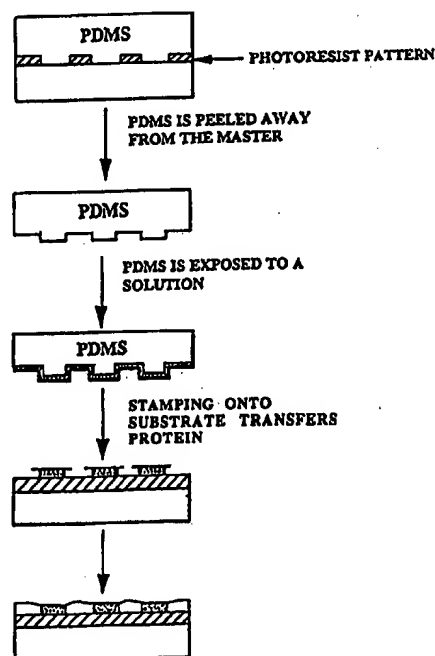
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(54) Title: PATTERNED DEPOSITION OF ANTIBODY BINDING PROTEINS FOR OPTICAL DIFFRACTION-BASED BIOSENSORS

(57) Abstract

The present invention provides an inexpensive and sensitive device and method for detecting and quantifying analytes present in a medium. The device comprises a metalized film upon which is printed a specific, predetermined pattern of an antibody-binding protein. Upon attachment of a target analyte to select areas of the plastic film upon which the protein is printed, diffraction of transmitted and/or reflected light occurs via the physical dimensions and defined, precise placement of the analyte. A diffraction image is produced which can be easily seen with the eye or, optionally, with a sensing device.



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10 **PATTERNED DEPOSITION OF ANTIBODY BINDING PROTEINS FOR
 OPTICAL DIFFRACTION-BASED BIOSENSORS**

TECHNICAL FIELD

15 The present invention is generally in the field of detecting analytes in a medium and, more particularly, the present invention relates to micro-contact printing of antibody-binding proteins onto a substrate for the development of single use, disposable sensors to indicate the presence of the analyte in a medium.

20 **BACKGROUND OF THE INVENTION**

 There are many systems and devices available for detecting a wide variety of analytes in various media. Most of these systems and devices are relatively expensive and require a trained technician to perform the test. There are many cases where it would be advantageous to be able to rapidly and inexpensively determine if an analyte were present. What is needed is a biosensor system that is easy and inexpensive to manufacture and is capable of reliable and sensitive detection of analytes, including smaller analytes. Additionally, what is needed is an easy flexible method of preparation of the biosensors which would permit optimum scale-up processing.

30 Sandstrom et al., 24 *Applied Optics* 472, 1985, describe use of an optical substrate of silicon with a layer of silicon monoxide and a layer of silicon formed as dielectric films. They indicate that a change in film thickness changes the properties of the optical substrate to produce different colors related to the thickness of the film. The thickness of the film is related to the color observed and a film provided on top of an optical substrate may produce a visible color change. The authors indicate that a mathematical model can be used to quantitate the color change, and that "[c]alculations

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performed using the computer model show that very little can be gained in optical performance from using a multilayer structure... but a biolayer on the surface changes the reflection of such structures very little since the optical properties are determined mainly by the interfaces inside the multilayer structure. The most sensitive system for detection of biolayers is a single layer coating, while in most other applications performance can be by additional dielectric layers."

Sandstrom et al., go on to indicate that slides formed from metal oxides on metal have certain drawbacks, and that the presence of metal ions can also be harmful in many biochemical applications. They indicate that the ideal top dielectric film is a 2-3 nm thickness of silicon dioxide which is formed spontaneously when silicon monoxide layer is deposited in ambient atmosphere, and that a 70-95 nm layer silicon dioxide on a 40-60 nm layer of silicon monoxide can be used on a glass or plastic substrate. They also describe formation of a wedge of silicon monoxide by selective etching of the silicon monoxide, treatment of the silicon dioxide surface with dichlorodimethylsilane, and application of a biolayer of antigen and antibody. From this wedge construction they were able to determine film thickness with an ellipsometer, and note that the "maximum contrast was found in the region about 65 nm where the interference color changed from purple to blue." They indicate that the sensitivity of such a system is high enough for the detection of protein antigen by immobilized antibodies. They conclude "the designs given are sensitive enough for a wide range of applications. The materials, i.e., glass, silicon, and silicon oxides, are chemically inert and do not affect the biochemical reaction studied. Using the computations above it is possible to design slides that are optimized for different applications. The slides can be manufactured and their quality ensured by industrial methods, and two designs are now commercially available.

U.S. Patent 5,512,131 issued to Kumar et al. describes a device that includes a polymer substrate having a metal coating. An antibody-binding protein layer is stamped on the coated substrate. The device is used in a process for stamping or as a switch. A diffraction pattern is generated when an analyte binds to the device. A visualization device, such as a spectrometer, is then used to determine the presence of the diffraction pattern.

However, the device described by Kumar et al. has several disadvantages. One disadvantage is that an extra visualization device is

needed to view any diffraction pattern. By requiring a visualization device, the Kumar et al. device does not allow a large number of samples to be tested since it is not possible to determine the presence of an analyte by using the unaided eye.

5 U.S. Patent No. 5,482,830 to Bogart, et al., describes a device that includes a substrate which has an optically active surface exhibiting a first color in response to light impinging thereon. This first color is defined as a spectral distribution of the emanating light. The substrate also exhibits a
10 second color which is different from the first color (by having a combination of wavelengths of light which differ from that combination present in the first color, or having a different spectral distribution, or by having an intensity of one or more of those wavelengths different from those present in the first color). The second color is exhibited in response to the same light when the analyte is present on the surface. The change from one color to another can
15 be measured either by use of an instrument, or by eye. Such sensitive detection is an advance over the devices described by Sandstrom and Nygren, supra, and allow use of the devices in commercially viable and competitive manner.

20 However, the method and device described in the Bogart, et al. patent has several disadvantages. One disadvantage is the high cost of the device. Another problem with the device is the difficulty in controlling the various layers that are placed on the wafer so that one obtains a reliable reading. What is needed is a biosensor device that is easy and inexpensive to manufacture and is capable of reliable and sensitive detection of the analyte to
25 be detected.

SUMMARY OF THE INVENTION

30 The present invention provides an inexpensive and sensitive device and method for detecting analytes present in a medium. The device comprises a biosensing device having a substrate, preferably a metalized polymer film, upon which is printed a specific predetermined pattern of antibody-binding proteins such as Protein A or Protein G. Subsequent exposure to the antibody specific for the desired analyte results in patterned deposition of this antibody. Overall, this allows a modular production format such that large
35 rolls of patterned protein may be made for use with different analytes. Then as needed, the final product may be made by exposure to the necessary antibody.

Upon attachment of a target analyte, which is capable of scattering light, to select areas of the polymer film upon which the protein and antibody are patterned, diffraction of transmitted and/or reflected light occurs via the physical dimensions and defined, precise placement of the analyte. A diffraction image is produced which can be easily seen with the eye or, optionally, with a sensing device.

The present invention utilizes methods of contact printing of patterned, antibody-binding proteins. These proteins bind to antibodies to pattern them on the surface as well as maintain the optimum orientation for the receptor antibodies. The receptor antibodies are specific for a particular analyte or class of analyte, depending upon the protein used. Methods of contact printing which would be useful in generating the sensing devices used in the present system are disclosed fully in U.S. Patent Application Nos. 08/707,456 and 08/769,594, both of which are incorporated herein by reference in their entirety. However, since these methods relate to self-assembling monolayers, the methods need to be altered slightly, as discussed below, to print the antibody-binding protein material as this material is not self-assembling.

Patterned antibody-binding protein layers with bound antibodies cause patterned placement or binding of analytes thereon. The biosensing devices of the present invention produced thereby may be used in one of two ways, depending on the size of the analyte. For analytes which are capable of causing diffraction by themselves, such as microorganisms, the system is used by first exposing the biosensing device to a medium that contains the analyte of choice and then, after an appropriate incubation period, transmitting a light, such as a laser, through the film or reflecting it off of the film. If the analyte is present in the medium and is bound to the antibodies on the patterned antibody-binding protein layer, the light is diffracted in such a way as to produce a visible image.

Optionally, for very small analytes such as proteins, the system may utilize "diffraction enhancing elements" which are capable of binding to the target analyte and to the biosensor and are capable of producing a substantial change in the height and/or refractive index, thereby increasing the diffraction efficiency of the biosensor and permitting the detection of smaller analytes. In use, a target analyte attaches either to the diffraction enhancing element, which then attaches to the biosensor, or directly to select areas of the polymer film upon which the protein and antibody are printed. Then

diffraction of transmitted and/or reflected light occurs via the physical dimensions and defined, precise placement of the analyte. A diffraction image is produced which can be easily seen with the eye or, optionally, with a sensing device.

5 Another option for use of this sensor involves the detection of analytes which are antibodies. The sensing device could comprise only the patterned antibody-binding proteins, and then would be exposed to the medium plus diffraction enhancing particles which have an antibody specific to the antibody to be detected. The selection of the antibody on the particle is
10 preferably made so that it does not bind non-specifically to the patterned antibody-binding protein, but instead binds only when the analyte antibody is also bound. In this way, the diffraction enhancing elements would cause a substantial change in the height and/or refractive index if the analyte antibody is present, thereby causing a diffraction image to form. It is envisioned that
15 the same format could be used with other immunoassay formats, such as lateral flow assays or microwell plates."

 In other words, the antibody-binding protein and antibody layers with the analyte bound thereto can produce optical diffraction patterns to indicate the presence of the analyte. The light can be in the visible spectrum, and be
20 either reflected from the film, or transmitted through it, and the analyte can be any compound or particle reacting with the antibody-binding protein layer. The light can be a white light or monochromatic electromagnetic radiation in the visible region. The present invention also provides a flexible support for an antibody-binding protein layer on gold or other suitable metal or metal
25 alloy.

 The present invention provides a low-cost, disposable biosensor which can be mass produced. It includes the use of surfaces patterned with an antibody-binding protein. Typically, these proteins bind an antibody by its constant region (F_c) so that the antibody's antigen-binding regions (F_{ab}) are
30 free for optimum binding activity. The preparation of the patterned protein surfaces also allows maximum flexibility in the sensor production. The final step in production, capturing the desired antibody in the patterned areas, may be done as needed for the desired analyte (i.e. at the time of manufacture).

 The biosensors of the present invention can be produced as a single
35 test for detecting an analyte or it can be formatted as a multiple test device. The biosensors of the present invention can be used to detect contamination in garments, such as diapers, and to detect contamination by microorganisms.

The present invention can also be used on contact lenses, eyeglasses, window panes, pharmaceutical vials, solvent containers, water bottles, adhesive bandages, and the like to detect contamination.

5 These and other features and advantages of the present invention will become apparent after a review of the following detailed description of the disclosed embodiments.

BRIEF DESCRIPTION OF THE FIGURES

10 Figure 1 shows a biosensor capable of simultaneously measuring several different analytes in a medium.

Figure 2 is a schematic of contact printing of antibody-binding protein layers, followed by patterned deposition of an antibody.

DETAILED DESCRIPTION

15 The present invention features improved biosensing devices, and methods for using such biosensing devices, for detecting and quantifying the presence or amount of an analyte of interest within a medium. The analytes that can be detected by the present invention include, but are not limited to, microorganisms such as bacteria, yeasts, fungi and viruses. In contrast to
20 prior devices, those of the present invention allow detection of extremely small quantities of analyte in a medium in a rapid assay lasting only a few minutes. In addition, no signaling or associated electronic components are required in the biosensing devices of the present invention.

25 The present invention comprises micro-contact printing of antibody-binding proteins onto a polymer film, which may also have a metal coating. This allows an easy, modular method of production in that subsequent exposure to an antibody causes its patterned binding. In addition, these proteins may enhance the activity of the immobilized antibody. The present invention allows for the development of single use, disposable biosensors
30 based on light diffraction to indicate the presence of the analyte. Upon attachment of a target analyte to select areas of the plastic film which contain the protein, diffraction of transmitted and/or reflected light occurs via the physical dimensions and defined, precise placement of the analyte. For example, yeast, fungi or bacterium are large enough to act as diffraction
35 elements for visible light when placed in organized patterns on a surface. Additionally, the present invention may include diffraction enhancing elements which increase the diffraction efficiency of the biosensor, thereby

making it possible to detect any number of different analytes. In addition to producing a simple diffraction image, patterns of analytes can be such as to allow for the development of a holographic sensing image and/or a change in visible color. Thus, the appearance of a hologram or a change in an existing
5 hologram will indicate a positive response. The pattern made by the diffraction of the transmitted light can be any shape including, but not limited to, the transformation of a pattern from one pattern to another upon binding of the analyte to the receptive material. In particularly preferred
10 embodiments, the diffraction pattern is discernible in less than one hour after contact of the analyte with the biosensing device of the present invention.

The diffraction grating which produces the diffraction of light upon interaction with the analyte preferably has a minimum periodicity of the wavelength and a refractive index different from that of the surrounding
15 medium. Very small analytes, such as viruses or molecules, can be detected indirectly by using a larger particle that is specific for the small analyte. In one embodiment in which the small analyte can be detected comprises coating the particle, such as a latex bead, with a protein material that specifically binds to the analyte of interest. Particles that can be used in the
20 present invention include, but are not limited to, glass, cellulose, synthetic polymers or plastics, latex, polystyrene, polycarbonate, proteins, bacterial or fungal cells and the like. The particles are preferably spherical in shape, but the structural and spatial configuration of the particles is not critical to the present invention. For instance, the particles could be slivers, ellipsoids,
25 cubes, and the like. A desirable particle size ranges from a diameter of approximately 0.2 μm to 50.0 μm , desirably between approximately 0.4 μm to 1 μm . The composition of the particle is not critical to the present invention.

The antibody which is immobilized/patterned on the surface will
30 specifically bind to an epitope on the analyte that is different from the epitope used in the binding to the diffraction enhancing element. Thus, for detecting a medium with a small analyte, such as viral particles, the medium is first exposed to the diffraction enhancing element particles, such as latex particles, to which the viral particles bind. Then, the diffraction enhancing element
35 particles are optionally washed and exposed to the polymer film with the antibody-binding protein layers containing the virus specific antibodies. The antibodies then bind to the viral particles on the element particle thereby

immobilizing the element particles in the same pattern as the antibodies on the film. Because the bound element particles will cause diffraction of the visible light, a diffraction pattern is formed, indicating the presence of the viral particle in the liquid. Additionally, the polymer film may include a metal coating thereon. The antibody-binding protein layer would then be located on the metalized surface of the film.

Alternatively, the analyte may be detected by first exposing the substrate to the medium containing the analyte and causing the analyte to bind to the antibody-binding protein layer material containing the analyte-specific antibody. Next, a solution containing the diffraction enhancing element particles is contacted with the substrate having the analyte bound thereto. The particles then bind to the analyte. Because the bound element particles will cause diffraction of the visible light, a diffraction pattern is formed, indicating the presence of the analyte in the liquid.

Finally, in a preferred embodiment, the biosensor, the diffraction enhancing element particles and the medium containing the analyte may be admixed simultaneously. This will result in a combination of the binding procedures discussed above. Some of the analytes will first bond with a diffraction enhancing element particle prior to binding to the substrate. Other analytes will first bind with the substrate and then bind with an element particle. When a point-light source is shown through the sensor, a diffraction pattern is formed, indicating the presence of the analyte in the liquid.

The analytes that are contemplated as being detected using the present invention include, but are not limited to, bacteria; yeasts; fungi; viruses; rheumatoid factor; antibodies, including, but not limited to IgG, IgM, IgA and IgE antibodies; carcinoembryonic antigen; streptococcus Group A antigen; viral antigens; antigens associated with autoimmune disease; allergens; tumor antigens; streptococcus Group B antigen; HIV I or HIV II antigen; or host response (antibodies) to these and other viruses; antigens specific to RSV or host response (antibodies) to the virus; an antigen; enzyme; hormone; polysaccharide; protein; lipid; carbohydrate; drug or nucleic acid; *Salmonella species*; *Candida species*, including, but not limited to *Candida albicans* and *Candida tropicalis*; *Salmonella species*; *Neisseria meningitides* groups A, B, C, Y and W sub 135, *Streptococcus pneumoniae*, *E. coli* K1, *Haemophilus influenza* type B; an antigen derived from microorganisms; a hapten, a drug of abuse; a therapeutic drug; an environmental agent; and antigens specific to hepatitis.

5 In another embodiment of the present invention, nutrients for a specific class of microorganisms can be incorporated into the antibody-binding protein layer. In this way, very low concentrations of microorganisms can be detected by first contacting the biosensor of the present invention with the nutrients incorporated therein and then incubating the biosensor under conditions appropriate for the growth of the bound microorganism. The microorganism is allowed to grow until there are enough organisms to form a diffraction pattern. Of course, in some cases, the microorganism can multiply enough to form a diffraction pattern without the presence of a nutrient on the patterned monolayer.

10 A part of the present invention is the method used to pattern receptors such as antibodies onto a polymer film or a metalized polymer film. The method entails microcontact printing proteins that bind to antibodies. These proteins can include, but are not limited to, Protein A, Protein G, Protein L, as well as their recombinant forms. Commercial versions, such as Zymed's (San Francisco, CA) KAPPA LOCKTM, are also suitable. Thus, the protein material is defined as the base material to create a specific binding pair, with the other part comprising an antibody specific to the analyte of interest.

20 The receptor material that is bound to the patterned protein is characterized by an ability to specifically bind the analyte or analytes of interest. Whatever the selected analyte of interest is, the protein material is designed to bind specifically with the analyte of interest. In the preferred embodiments, the biosensing device is configured and arranged to provide a pattern detectable by eye in response to transmission of polychromatic light when the analyte of interest is sandwiched between the antibody and a diffraction enhancing element. In another embodiment, where the analyte is large enough to diffract light, no diffraction enhancing element may be needed.

25 In many instances, a "blocker" may be necessary to prevent non-specific binding. The term "blocker" as used herein means a reagent that adheres to the sensor surface so that it "blocks" or prevents non-analyte materials from non-specifically binding to the surface (either in the patterned or un-patterned areas). The blocking step may be done as a post-treatment to a surface which has already been contact printed ("post-block"), and is the standard technique for filling in non-contact printed regions with another thiol. However, the inventors have discovered that a "pre-block" technique is preferred over the post-block technique. In the pre-block technique, the

5 surface of the substrate is pre-treated with a non-thiol containing blocker and then contact printed. Not wishing to be bound to any theory, it is theorized that the contact printed material (usually sulfur containing) displaces the physisorbed blocker, thereby permitting the antibody-binding protein material to be bound directly to the surface of the substrate. A subsequent post-block may also be performed, if desired. Blockers can include, but are not limited to, β -casein, albumins such as bovine serum albumin, pluronic or other surfactants, polyethylene glycol, polyvinyl alcohol, or sulfur derivatives of the above compounds, and any other blocking material known to those of
10 ordinary skill in the art.

The matrix containing the analyte of interest may be a solid, a gas, or a bodily fluid such as an interstitial fluid, mucous, saliva, urine, fecal material, tissue, marrow, cerebral spinal fluid, serum, plasma, whole blood, synovial sputum, buffered solutions, extracted solutions, semen, vaginal secretions,
15 pericardial, gastric, peritoneal, pleural, a throat swab or other washes and the like. The analyte of interest may be an antigen, an antibody, an enzyme, a toxin, an environmental agent, a cytoplasm component, pili or flagella component, protein, polysaccharide, drug, or any other material that is capable of being recognized by an antibody. For example, receptor material for bacteria may specifically bind a surface membrane component, protein or
20 lipid, a polysaccharide, a nucleic acid, or an enzyme. The analyte which is indicative of the bacteria may be a saccharide or polysaccharide, an enzyme, a nucleic acid, a membrane component, a ganglioside or an antibody produced by the host in response to the bacteria. The presence of the analyte may indicate an infectious disease (bacterial or viral), cancer, an allergy, or
25 other medical disorder or condition. The presence of the analyte may be an indication of water or food contamination or other harmful materials. The analyte may indicate drug abuse or may monitor levels of therapeutic agents.

In some cases, the analyte may not simply bind the receptor material,
30 but may cause a detectable modification of the receptor material to occur. This interaction could cause an increase in mass at the test surface or a decrease in the amount of receptor material on the test surface. An example of the latter is the interaction of a degradative enzyme or material with a specific, immobilized substrate. In this case, one would see a diffraction pattern before interaction with the analyte of interest, but the diffraction
35 pattern would disappear if the analyte were present. The specific mechanism through which binding, hybridization, or interaction of the analyte with the

receptor material occurs is not important to this invention, but may impact the reaction conditions used in the final assay protocol.

In general, the antibody-binding protein may be passively adhered to the substrate layer. If required, functional groups may be conjugated onto the antibody-binding protein to allow its covalent attachment onto the test surface.

A wide range of techniques can be used to apply the protein material to the substrate. Test surfaces may be coated with antibody-binding protein by application of solution in discrete arrays or patterns; spraying, ink jet, or other imprinting methods; or by contact printing. The technique selected should minimize the amount of protein material required for coating a large number of test surfaces and maintain the stability/functionality of protein material during application. The technique must also apply or adhere the protein material to the substrate in a very uniform and reproducible fashion.

The next step would comprise exposing the patterned surface to the antibody specific for the desired analyte. This could be done by total immersion in a solution for a predetermined period of time, spraying, or spin coating. In addition, controlled placement of various antibodies could lead to a multi-analyte system. Another advantage to this process is the modular format for production. In other words, one system with patterned antibody-binding protein can be optimized and then provide detection of a wide variety of analytes, depending on the antibody bound.

The medium in which the analyte may reside can be solid, gel-like, liquid or gas. For purposes of detecting an analyte in a body fluid, the fluid is selected from, but not limited to, urine, serum, plasma, spinal fluid, sputum, whole blood, saliva, uro-genital secretions, fecal extracts, pericardial, gastric, peritoneal, pleural washes, vaginal secretions, or a throat swab; and the method optionally includes using a spectrophotometer to measure the appearance of the refractive pattern. The most common gas that is contemplated as being used with the biosensing device of the present invention is air.

The biosensing device of the present invention utilizes methods of contact printing of patterned, antibody-binding proteins on substrates, desirably metalized polymer films, the subsequent exposure to antibody to achieve patterned antibody, the compositions produced thereby, and the use of these compositions. Patterned antibody-binding protein layers allow for the controlled placement of antibodies thereon which can bind an analyte.

The term "patterned antibody-binding protein layers" as used herein means the antibody-binding protein plus the desired antibody layers in any pattern on the substrates including a solid pattern.

5 When the film with the patterned antibody-binding protein layers is exposed to an analyte that is capable of reacting with the antibody, the film will produce optical diffraction patterns which differ depending on the reaction of the antibody with the analyte of interest. The liquid may be a high surface tension fluid such as water. The light can be in the visible spectrum, and be either reflected from the film, or transmitted through it, and
10 the analyte can be any compound reacting with the patterned antibody-binding protein layer.

In preferred embodiments, the method involves contacting the substrate with a test sample potentially containing the analyte under conditions in which the substrate causes a change in the refractive index.
15 When light is transmitted through the film with the patterned antibody-binding protein layer, a visible diffraction pattern is formed and can be visualized by directing the light to a surface or by looking directly through the substrate.

In one embodiment, the present invention is contemplated in a dipstick form in which the micro-contact printed film is mounted at the end of the dipstick. In use the dipstick is dipped into the liquid in which the suspected analyte may be present and allowed to remain for several minutes. The dipstick is then removed and then, either a light is projected through the film or the film is observed with a light behind the film. If a pattern is observed,
20 then the analyte is present in the liquid.

In another embodiment of the present invention, a multiple analyte test is constructed on the same support. As shown in Figure 1, a strip 10 is provided with several micro-contact printed films 20, 25, 30 and 35, each film having a pattern 40 printed thereon. Each of the micro-contact printed
25 metalized films 15, 20, 25, and 30 have a different antibody that is specific for different analytes. It can be seen that the present invention can be formatted in any array with a variety of micro-contact printed metalized films thereby allowing the user of the biosensor device of the present invention to detect the presence of multiple analytes in a medium using a single test.

35 In yet another embodiment of the present invention, the biosensor can be attached to an adhesively backed sticker or decal which can then be placed on a hard surface or container wall. The biosensor can be placed on the

inside surface of a container such as a food package or a glass vial. The biosensor can then be visualized to determine the presence of analyte.

Typically, a gold film, 5 to 2000 nm thick, is supported on a titanium-primed polyethylene-terephthalate film, Si/SiO₂ wafer or glass sheet.

5 The titanium serves as an adhesion promoter between gold and the support. The antibody-binding protein attaches to the gold surface during contact printing.

10 Figure 2 outlines the procedure used for microcontact printing. An elastomeric stamp is used to transfer antibody-binding protein "ink" to a surface by contact; if the stamp is patterned, a patterned antibody-binding protein layer forms. The stamp is fabricated by casting polydimethylsiloxane (PDMS) on a master having the desired pattern. Masters are prepared using standard photolithographic techniques, or constructed from existing materials having microscale surface features.

15 In a preferred embodiment of a typical experimental procedure, a photolithographically produced master is placed in a glass or plastic Petri dish, and a 10:1 ratio (w:w) mixture of SYLGARD® silicone elastomer 184 and SYLGARD® silicone elastomer 184 curing agent (Dow Corning Corporation) is poured over it. The elastomer is allowed to sit for
20 approximately 30 minutes at room temperature and reduced pressure to degas, then cured for at least 4 hours at 60°C, and gently peeled from the master. "Inking" of the elastomeric stamp is accomplished by exposing the stamp to a 0.1 to 10 µM aqueous solution of disulfide-derivatized antibody-binding protein typically by placing the stamp face down in the solution for
25 10 seconds to 10 minutes. The stamp is allowed to dry, either under ambient conditions, or typically by exposure to a stream of air or nitrogen gas. Following inking, the stamp is applied to a gold surface. Light pressure is used to ensure complete contact between the stamp and the surface. After 1 second to 5 minutes, the stamp is then gently peeled from the surface.
30 Following removal of the stamp, the surface is rinsed and dried. Alternatively, further derivatization of unstamped areas can be accomplished, either by using a second stamp or by exposing the entire surface with a different reagent. Subsequently, exposure to a protein-blocking agent, such as BSA or β-casein, or any other agent well known in the art, can also be
35 done. After the pattern is positioned, the patterned surface would be exposed to an antibody specific for the desired analyte either by immersion in a

solution or by spraying or spin coating the solution onto the patterned surface.

5 The elastomeric character of the stamp is important to the success of the process. Polydimethylsiloxane (PDMS), when cured, is sufficiently elastomeric to allow good conformal contact of the stamp and the surface, even for surfaces with significant relief; this contact is essential for efficient contact transfer of the protein to the gold film. The elastomeric properties of PDMS are also important when the stamp is removed from the master: if the stamp were rigid (as is the master) it would be difficult to separate the stamp and master after curing without damaging one of the two substrates. PDMS is also sufficiently rigid to retain its shape, even for features with sub-micron dimensions. The surface of PDMS has a low interfacial free energy ($\gamma = 22.1$ dynes/cm), and the stamp does not adhere to the gold film. The stamp is durable in that the same stamp can be used up to 100 times over a period of several months without significant degradation in performance. The polymeric nature of PDMS also plays a critical role in the inking procedure, by enabling the stamp to absorb the protein ink by swelling.

10 A more detailed description of the methods and compositions of the present invention follows. All publications cited herein are incorporated by reference in their entirety.

20 Any plastic film is suitable for the present invention. Preferably, the plastic film is also capable of having a metal coating deposited thereon. These include, but are not limited to polymers such as: polyethylene-terephthalate (MYLAR®), acrylonitrile-butadiene-styrene, acrylonitrile-methyl acrylate copolymer, cellophane, cellulosic polymers such as ethyl cellulose, cellulose acetate, cellulose acetate butyrate, cellulose propionate, cellulose triacetate, cellulose triacetate, polyethylene, polyethylene - vinyl acetate copolymers, ionomers (ethylene polymers) polyethylene-nylon copolymers, polypropylene, methyl pentene polymers, polyvinyl fluoride, and aromatic polysulfones. Preferably, the plastic film has an optical transparency of greater than 80%. Other suitable thermoplastics and suppliers may be found, for example, in reference works such as the *Modern Plastics Encyclopedia* (McGraw-Hill Publishing Co., New York 1923-1996).

30 In one embodiment of the invention, the polymer film has a metal coating thereon and has an optical transparency of between approximately 5% and 95%. A more desired optical transparency for the thermoplastic film used in the present invention is between approximately 20% and 80%. In a

desired embodiment of the present invention, the polymer film has at least an approximately 80% optical transparency, and the thickness of the metal coating is such as to maintain an optical transparency greater than about 20%, so that diffraction patterns can be produced by either reflected or transmitted light. This corresponds to a metal coating thickness of about 10 nm. However, in other embodiments of the invention, the gold thickness may be between approximately 1 nm and 1000 nm.

The preferred metal for deposition on the film is gold. However, silver, aluminum, chromium, copper, iron, zirconium, platinum and nickel, as well as oxides of these metals, may be used.

In principle, any surface with corrugations of appropriate size could be used as masters. The process of microcontact printing starts with an appropriate relief structure, from which an elastomeric stamp is cast. This 'master' template may be generated photolithographically, or by other procedures, such as commercially available diffraction gratings. In one embodiment, the stamp may be made from polydimethylsiloxane.

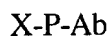
In another embodiment, the invention features an optical assay device, having an optically active receptive surface configured and arranged to allow simultaneous assay of a plurality of samples on the surface for one analyte of interest, and an automated liquid handling apparatus (e.g., a pipetting device) configured and arranged to dispense sample and reagent solutions to the surface.

Below is provided an indication of the methodology by which the optimal materials and methods useful for construction of optical test surfaces of this invention can be made. Generally, the present invention includes novel optically active test surfaces for the direct detection of an analyte. These test surfaces have an analyte-specific antibody bound to the test surface by use of an attachment layer – namely the antibody-binding proteins. Thus, the present invention provides a detection device which includes selecting an optical substrate, patterning it with antibody-binding protein, and then exposing this to the antibody for the desired analyte. The detection method involves contacting this device with a sample fluid containing the analyte of interest, and then examining the change in diffraction of transmitted or reflected light by observing whether a diffraction pattern is formed.

The present invention has a broad range of applications and may be utilized in a variety of specific binding pair assay methods. For example, the devices of this invention can be used in immunoassay methods for either

antigen or antibody detection. The devices may be adapted for use in direct, indirect, or competitive detection schemes.

In one embodiment of the present invention, the antibody-binding protein layer has the following general formula:



X is optional as a means of allowing chemisorption to a metal or metal oxide. For example, X may be asymmetrical or symmetrical disulfide (-R'SSY', -RSSY), sulfide (-R'SY', -RSY), diselenide (-R'Se-SeY'), selenide (-R'SeY', -RSeY), thiol (-SH), nitrile (-CN), isonitrile, nitro (-NO₂), selenol (-SeH), trivalent phosphorous compounds, isothiocyanate, xanthate, thiocarbamate, phosphine, thioacid or dithioacid, carboxylic acids, hydroxylic acids, and hydroxamic acids.

P represents the antibody-binding proteins which may be derivatized with X. Ab represents the antibody specific to the desired analyte.

The stamp may be applied in air, or under a fluid such as water to prevent excess diffusion of the protein material. For large-scale or continuous printing processes, it is most desirable to print in air, since shorter contact times are desirable for those processes.

In one embodiment of the present invention, the pattern is formed on the metalized thermoplastic polymer with the antibody-binding protein layer. In another embodiment of the present invention, the relief of the pattern is formed with the antibody-binding protein layer. After the stamping process, the metalized areas on the plastic may optionally be passivated or blocked, for example, with a reagent such as β -casein. Preferably this is done prior to exposure to the antibody.

This invention is further illustrated by the following examples, which are not to be construed in any way as imposing limitations upon the scope thereof. On the contrary, it is to be clearly understood that resort may be had to various other embodiments, modifications, and equivalents thereof, which, after reading the description herein, may suggest themselves to those skilled in the art without departing from the spirit of the present invention.

EXAMPLES

EXAMPLE 1

5 Antibody-conjugated polystyrene particles were produced by carbodiimide coupling with ethyldimethylaminodicarbodiimide (EDAC, bottle No. 3 of Polysciences kit, Catalog No. 19539). For this example, 0.125 mL of a 10% suspension of 0.5 micron diameter blue carboxylated particles (Bangs Laboratories; Fishers, Indiana; Cat #D0005070CB) were activated with an aqueous solution of the EDAC for 1-4 hours, rinsed, then exposed to 300 micrograms of a monoclonal antibody to luteinizing hormone, alpha subunit, (Fitzgerald Industries, Cat. No. 10-L10, Clone No. M94136). The particles were again rinsed, blocked with bovine serum albumin, and stored at 2.5% concentration in phosphate buffered saline.

10 A 10 mM aqueous stock solution of Sulfo-LC-SPDP (Pierce Chemical Co.; Rockford, IL) was prepared by dissolving 1.3 mg Sulfo-LC-SPDP into 2.07 mL de-ionized water. The conjugation reaction was carried out in phosphate buffered saline (PBS) containing 20 mM sodium phosphate buffer, 150 mM NaCl, 1 mM EDTA, and 0.02% sodium azide at pH 7.5. One milligram of lyophilized Protein A or Protein G was dissolved in 450 microliters PBS, and 50 microliters of Sulfo-LC-SPDP stock solution was added to the antibody solution. The mixture was allowed to react at room temperature for 60 minutes. The sample was applied to a 5-mL desalting polyacrylamide column previously equilibrated with 5 bed volumes (25 mL) of PBS. Fractions were eluted using PBS as the elution buffer, and the protein in the fractions was monitored using a COOMASSIE[®] Plus Protein Assay (Pierce Chemical Co.). Typically, 50 μ L of the COOMASSIE[®] reagent was mixed with 50 μ L of each fraction in a micro-titer plate. The COOMASSIE[®] reagent reacted with the protein, producing a blue color, the intensity of which was dependent upon the amount of protein present in the fraction. The fractions which produced the most intense blue color were those containing the majority of the protein eluted. These fractions were pooled together as the disulfide form of the final derivatized product. This was typically the form used for contact printing.

30 Optionally, the disulfide-pyridyl group present on the disulfide form of the thiolated binder can be reduced to a thiol group in a reduction reaction. Instead of desalting on a column equilibrated with PBS, the derivatized protein was desalted on a column equilibrated with an acetate buffer (100mM

sodium acetate buffer, 100 mM NaCl, pH 4.5). The acidic pH of this acetate buffer acts to protect any disulfide bonds present on the native protein from undesired reduction. In the reduction reaction, 12 mg of dithiothreitol (DTT) is dissolved in 500 mL acetate buffer and added to 1 mL of the SPDP derivatized protein. The reaction mixture was incubated for 30 minutes at room temperature, and desalted on a 5 mL desalting column equilibrated with 5 bed volumes (25 mL) of acetate buffer. The protein content of the fractions eluted was again monitored by the COOMASSIE[®] Protein Assay reagent as described above, and the fractions containing the greatest amount of protein were pooled for subsequent contact printing.

Both the disulfide and reduced forms of the thiolated binders were stored in aqueous solution at 4 °C until used for contact printing.

A gold/MYLAR[®] film was pre-treated (or blocked) with a 5 mg/mL beta casein solution for 10 minutes, then thoroughly rinsed and dried under an air stream. A PDMS stamp of 10-micron circles was coated with thiolated antibody-binding protein by placing the stamp face down in a 0.5 mg/mL thiolated Protein A (or Protein G) solution and soaking for 10 minutes. A strong air stream was used to thoroughly dry the surface of the stamp. The coated stamp is placed in contact with the gold/MYLAR[®] for 5 minutes, then removed. The resulting printed gold/MYLAR[®] was rinsed in distilled water, and dried.

The sensor was then exposed to an antibody solution (e.g., a 2 ug/mL PBS solution of monoclonal antibody to luteinizing hormone-beta, Catalog No. 10-L15, Clone No. M94187, Fitzgerald Industries International, Inc.; Concord, MA) for 30 minutes, followed by rinsing, and air-drying.

The sensor sample was exposed to a 2 ug/mL aqueous solution (with 1% bovine serum albumin) of goat anti-mouse HRP (horse radish peroxidase labeled) by placing a droplet of the solution on top of the sensor surface for 30-60 minutes at room temperature. The sample was rinsed with 0.02% Tween 20 solution, then distilled water. A subsequent exposure to a TMB membrane enhancer solution (e.g., a 10:1 v/v mixture of Kirkegaard and Perry Laboratories' reagents Cat No. 50-76-18 and Cat. No. 50-77-01) was done by placing the TMB solution on the sensor sample for 10 minutes. This caused the development of a blue precipitate in the circles or features, as well as a diffraction image to form upon irradiation with a point light source.

Optionally, the analyte solution (e.g., luteinizing hormone for this example) is mixed with microparticles (typically 50-70 microliters of analyte

5 solution with 10-20 microliters of 1.5-2.5% antibody-conjugated particle suspension), and placed on top of the sensor (typically a 1 cm square sensor sample is used). After 5-10 minutes, a nitrocellulose disk (5 or 8 micron pore size, Sigma No. N3771 or N4146) with a small hole punched out of the center is placed on top of the sensor. This acts to wick away excess fluid and unbound microparticles. At that time, a point light source is transmitted through the sensor sample (using the small hole in the nitrocellulose). If the analyte was present, then a diffraction image is seen on the other side of the light beam.

10 EXAMPLE 2

Antibody-conjugated polystyrene particles were produced by carbodiimide coupling with ethyldimethylaminodicarbodiimide (EDAC, bottle No. 3 of Polysciences kit, Catalog No. 19539). For this example, 0.125 mL of a 10% suspension of 0.5 micron diameter blue carboxylated particles (Bangs Laboratories; Fishers, Indiana; Cat. No. D0005070CB) were activated with an aqueous solution of the EDAC for 1-4 hours, rinsed, then exposed to 300 micrograms of a polyclonal antibody to IgE (such as chicken anti-IgE in order to be unreactive to the patterned Protein A). The particles were again rinsed, blocked with bovine serum albumin, and stored at 2.5% concentration in phosphate buffered saline.

20 A 10 mM aqueous stock solution of Sulfo-LC-SPDP (Pierce Chemical Co.; Rockford, IL) was prepared by dissolving 1.3 mg Sulfo-LC-SPDP into 2.07 mL de-ionized water. The conjugation reaction was carried out in phosphate buffered saline (PBS) containing 20 mM sodium phosphate buffer, 150 mM NaCl, 1 mM EDTA, and 0.02% sodium azide at pH 7.5. One milligram of lyophilized Protein A was dissolved in 450 microliters PBS, and 50 microliters of Sulfo-LC-SPDP stock solution was added to the antibody solution. The mixture was allowed to react at room temperature for 60 minutes. The sample was applied to a 5-mL desalting polyacrylamide column previously equilibrated with 5 bed volumes (25 mL) of PBS. Fractions were eluted using PBS as the elution buffer, and the protein in the fractions were monitored using a COOMASSIE[®] Plus Protein Assay (Pierce Chemical Co.). Typically, 50 μ L of the COOMASSIE[®] reagent was mixed with 50 μ L of each fraction in a micro-titer plate. The COOMASSIE[®] reagent reacted with the protein, producing a blue color, the intensity of which was dependent upon the amount of protein present in the fraction. The fractions which

produced the most intense blue color were those containing the majority of the protein eluted. These fractions were pooled together as the disulfide form of the final derivatized product. This was typically the form used for contact printing.

5 Optionally, the disulfide-pyridyl group present on the disulfide form of the thiolated binder can be reduced to a thiol group in a reduction reaction. Instead of desalting on a column equilibrated with PBS, the derivatized protein was desalted on a column equilibrated with an acetate buffer (100mM sodium acetate buffer, 100 mM NaCl, pH 4.5). The acidic pH of this acetate
10 buffer acts to protect any disulfide bonds present on the native protein from undesired reduction. In the reduction reaction, 12 mg of dithiothreitol (DTT) was dissolved in 500 mL acetate buffer and added to 1 mL of the SPDP derivatized protein. The reaction mixture was incubated for 30 minutes at room temperature, and desalted on a 5 mL desalting column equilibrated
15 with 5 bed volumes (25 mL) of acetate buffer. The protein content of the fractions eluted was again monitored by the COOMASSIE[®] Protein Assay reagent as described above, and the fractions containing the greatest amount of protein were pooled for subsequent contact printing.

20 Both the disulfide and reduced forms of the thiolated binders were stored in aqueous solution at 4°C until used for contact printing.

25 A gold/MYLAR[®] film was pre-treated (or blocked) with a 5 mg/mL beta casein solution for 10 minutes, then thoroughly rinsed and dried under an air stream. A PDMS stamp of 10-micron circles was coated with thiolated antibody-binding protein by placing the stamp face down in a 0.5 mg/mL thiolated Protein A solution and soaking for 10 minutes. A strong air stream was used to thoroughly dry the surface of the stamp. The coated stamp was placed in contact with the gold/MYLAR[®] for 5 minutes, then removed. The resulting printed gold/MYLAR[®] was rinsed in distilled water, and dried.

30 The analyte solution (typically 50-70 microliters at 2 µg/mL PBS solution of IgE (Catalog No. 30-AI05, Fitzgerald Industries International, Inc.; Concord, MA) is placed on top of the sensor (typically a 1 cm square) for 0-20 minutes, followed by 10-20 microliters of 1.5-2.5% antibody-conjugated particle suspension for an additional 5-20 minutes. A nitrocellulose disk (5 or 8 micron pore size, Sigma No. N3771 or N4146) with a small hole punched
35 out of the center is then placed on top of the sensor. This acts to wick away excess fluid and unbound microparticles. At that time, a point light source is transmitted through the sensor sample (using the small hole in the

nitrocellulose). If the analyte was present, then a diffraction image is seen on the other side of the light beam, as illustrated.

We claim:

1. A biosensor comprising:
a polymer film; and
5 an antibody-binding protein layer printed in a pattern onto the polymer film wherein the antibody-binding protein layer has an antibody thereon that is specific for an analyte.

10 2. The biosensor of Claim 1, wherein the antibody-binding protein layer is printed in a pattern such that when the biosensor binds the analyte, the biosensor diffracts transmitted light to form a diffraction pattern.

15 3. The biosensor of Claim 2, wherein the diffraction pattern is visible with an unaided eye.

4. The biosensor of Claim 1, wherein the polymer film further comprises a metal coating.

20 5. The biosensor of Claim 4, wherein the metal is selected from gold, silver, chromium, nickel, platinum, aluminum, iron, copper, zirconium, or oxides thereof.

6. The biosensor of Claim 4, wherein the metal is gold.

25 7. The biosensor of Claim 6, wherein the gold coating is between approximately 1 nanometer and 1000 nanometers in thickness.

30 8. The biosensor of Claim 1, wherein the polymer film is selected from polyethylene-terephthalate, acrylonitrile-butadiene-styrene, acrylonitrile-methyl acrylate copolymer, cellophane, cellulosic polymers such as ethyl cellulose, cellulose acetate, cellulose acetate butyrate, cellulose propionate, cellulose triacetate, cellulose triacetate, polyethylene, polyethylene - vinyl acetate copolymers, ionomers (ethylene polymers) polyethylene-nylon copolymers, polypropylene, methyl pentene polymers, polyvinyl fluoride, or
35 aromatic polysulfones.

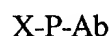
9. The biosensor of Claim 8, wherein the polymer film is polyethylene-terephthalate.

10. The biosensor of Claim 1, wherein the polymer film is optically transparent.

11. The biosensor of Claim 1, wherein the polymer film has an optical transparency between 5% and 95%.

12. The biosensor of Claim 1, wherein the polymer film has an optical transparency between approximately 20% and 80%.

13. The biosensor of Claim 1, wherein the antibody-binding protein layer is formed from compounds with the following general formula:



wherein:

X is reactive with the metal or metal oxide on the polymer film;

P is an antibody-binding protein; and

Ab is an antibody specific to a desired analyte.

14. The biosensor of Claim 13, wherein:

X is a asymmetrical or symmetrical disulfide (-SSY', -SSY), sulfide (-'SY', SY), diselenide (-'Se-SeY'), selenide (-SeY', -SeY), thiol (-SH), nitrile (-CN), isonitrile, nitro (-NO₂), selenol (-SeH), trivalent phosphorous compounds, isothiocyanate, xanthate, thiocarbamate, phosphine, thioacid or dithioacid, carboxylic acids, hydroxylic acids, and hydroxamic acids.

15. The biosensor of Claim 1, wherein the analyte is selected from bacteria, yeast, fungus, virus, rheumatoid factor, IgG, IgM, IgA and IgE antibodies, carcinoembryonic antigen, streptococcus Group A antigen, viral antigens, antigens associated with autoimmune disease, allergens, tumor
5 antigens, streptococcus Group B antigen, HIV I or HIV II antigen, antibodies, viruses, antigens specific to RSV, antigen, enzyme, hormone, polysaccharide, protein, lipid, carbohydrate, drug, nucleic acid, *Neisseria meningitides* groups A, B, C, Y and W sub 135, *Streptococcus pneumoniae*, *E. coli* K1, *Haemophilus influenza* type B, an antigen derived from microorganisms, a
10 hapten, a drug of abuse, a therapeutic drug, an environmental agent, or antigens specific to hepatitis.

16. The biosensor of Claim 15, wherein the analyte is bacteria, yeast, fungus or virus.

17. The biosensor of Claim 16, wherein the fungus is *Candida species*.

18. The biosensor of Claim 16, wherein the bacteria is *Salmonella species*.

19. The biosensor of Claim 1, wherein the protein material is selected from Protein A, Protein G, Protein L, or a recombinant form thereof.

20. A method of making a biosensor comprising printing a pattern of antibody-binding protein layer with a subsequent layer of antibody on the polymer film.

21. The method of Claim 20, wherein the antibody-binding protein layer is printed in a pattern such that when the biosensor binds an analyte, the biosensor diffracts transmitted light to form a diffraction pattern.

22. The method of Claim 20, wherein the polymer film further comprises a metal coating.

23. The method of Claim 22, wherein the metal is selected from gold, silver, chromium, nickel, platinum, aluminum, iron, copper, zirconium, or oxides thereof.

5 24. The method of Claim 22, wherein the metal is gold.

25. The method of Claim 24, wherein the gold coating is between approximately 1 nanometer and 1000 nanometers in thickness.

10 26. The method of Claim 20, wherein the polymer film is selected from polyethylene-terephthalate, acrylonitrile-butadiene-styrene, acrylonitrile-methyl acrylate copolymer, cellophane, cellulosic polymers such as ethyl cellulose, cellulose acetate, cellulose acetate butyrate, cellulose propionate, cellulose triacetate, cellulose triacetate, polyethylene, polyethylene - vinyl
15 acetate copolymers, ionomers (ethylene polymers) polyethylene-nylon copolymers, polypropylene, methyl pentene polymers, polyvinyl fluoride, or aromatic polysulfones.

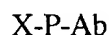
20 27. The method of Claim 26, wherein the polymer film is polyethylene-terephthalate.

28. The method of Claim 20, wherein the polymer film is optically transparent.

25 29. The method of Claim 28, wherein the polymer film has an optical transparency between 5% and 95%.

30 30. The method of Claim 28, wherein the polymer film has an optical transparency between approximately 20% and 80%.

31. The method of Claim 20, wherein the antibody-binding protein layer is formed from compounds with the following general formula:



wherein:

X is reactive with the metal or metal oxide on the polymer film;

P is an antibody-binding protein; and

Ab is an antibody specific to a desired analyte.

32. The method of Claim 31, wherein:

X is a asymmetrical or symmetrical disulfide (-SSY', -SSY), sulfide (-'SY', SY), diselenide (-'Se-SeY'), selenide (-SeY', -SeY), thiol (-SH), nitrile (-CN), isonitrile, nitro (-NO₂), selenol (-SeH), trivalent phosphorous compounds, isothiocyanate, xanthate, thiocarbamate, phosphine, thioacid or dithioacid, carboxylic acids, hydroxylic acids, and hydroxamic acids.

33. The method of Claim 20, wherein the analyte is selected from bacteria, yeast, fungus, virus, rheumatoid factor, IgG, IgM, IgA and IgE antibodies, carcinoembryonic antigen, streptococcus Group A antigen, viral antigens, antigens associated with autoimmune disease, allergens, tumor antigens, streptococcus Group B antigen, HIV I or HIV II antigen, antibodies, viruses, antigens specific to RSV, antigen, enzyme, hormone, polysaccharide, protein, lipid, carbohydrate, drug, nucleic acid, *Neisseria meningitides* groups A, B, C, Y and W sub 135, *Streptococcus pneumoniae*, *E. coli* K1, *Haemophilus influenza* type B, an antigen derived from microorganisms, a hapten, a drug of abuse, a therapeutic drug, an environmental agent, or antigens specific to hepatitis.

34. The method of Claim 33, wherein the analyte is bacteria, yeast, fungus or virus.

35. The method of Claim 20, wherein the protein material is selected from Protein A, Protein G, Protein L, or a recombinant form thereof.

36. A method of detecting an analyte in a medium comprising;
contacting the medium suspected of containing the analyte with a
biosensing device, the biosensing device comprising:

a polymer film; and
5 an antibody-binding protein layer printed in a pattern onto the polymer film
wherein the antibody-binding protein layer has a protein material thereon that
is specific for the analyte; and

transmitting a light through the polymer film; and
detecting presence of the analyte bound to the protein material by
10 detecting a pattern form by diffraction of the transmitted light.

37. The method of Claim 36, wherein the diffraction pattern is
visible with an unaided eye.

15 38. The method of Claim 36, wherein the polymer film further
comprises a metal coating.

39. The method of Claim 38, wherein the metal is gold.

20 40. A biosensor comprising:
a polymer film; and
an antibody-binding protein layer printed in a pattern onto the polymer
film wherein the antibody-binding protein layer is capable of acting as a
receptor for an analyte.

25 41. The biosensor of Claim 40, wherein the antibody-binding protein
layer is printed in a pattern such that when the biosensor binds the analyte,
the biosensor diffracts transmitted light to form a diffraction pattern.

30 42. The biosensor of Claim 41, wherein the diffraction pattern is
visible with an unaided eye.

43. The biosensor of Claim 40, wherein the polymer film further
comprises a metal coating.

44. The biosensor of Claim 43, wherein the metal is selected from gold, silver, chromium, nickel, platinum, aluminum, iron, copper, zirconium, or oxides thereof.

5 45. The biosensor of Claim 43, wherein the metal is gold.

46. The biosensor of Claim 45, wherein the gold coating is between approximately 1 nanometer and 1000 nanometers in thickness.

10 47. The biosensor of Claim 40, wherein the polymer film is selected from polyethylene-terephthalate, acrylonitrile-butadiene-styrene, acrylonitrile-methyl acrylate copolymer, cellophane, cellulosic polymers such as ethyl cellulose, cellulose acetate, cellulose acetate butyrate, cellulose propionate, cellulose triacetate, cellulose triacetate, polyethylene, polyethylene - vinyl
15 acetate copolymers, ionomers (ethylene polymers) polyethylene-nylon copolymers, polypropylene, methyl pentene polymers, polyvinyl fluoride, or aromatic polysulfones.

20 48. The biosensor of Claim 47, wherein the polymer film is polyethylene-terephthalate.

49. The biosensor of Claim 40, wherein the polymer film is optically transparent.

25 50. The biosensor of Claim 40, wherein the polymer film has an optical transparency between 5% and 95%.

30 51. The biosensor of Claim 40, wherein the polymer film has an optical transparency between approximately 20% and 80%.

52. The biosensor of Claim 40, wherein the antibody-binding protein layer is formed from compounds with the following general formula:



5

wherein:

X is reactive with the metal or metal oxide on the polymer film;

P is an antibody-binding protein; and

10

53. The biosensor of Claim 52, wherein:

X is a asymmetrical or symmetrical disulfide (-SSY', -SSY), sulfide (-'SY', SY), diselenide (-'Se-SeY'), selenide (-SeY', -SeY), thiol (-SH), nitrile (-CN), isonitrile, nitro (-NO₂), selenol (-SeH), trivalent phosphorous compounds, isothiocyanate, xanthate, thiocarbamate, phosphine, thioacid or dithioacid, carboxylic acids, hydroxylic acids, and hydroxamic acids.

15

54. The biosensor of Claim 40, wherein the analyte is selected from antibodies such as IgG, IgM, IgA and IgE.

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55. The biosensor of Claim 40, wherein the protein material is selected from Protein A, Protein G, Protein L, or a recombinant form thereof.

25

56. The biosensor of Claim 40, further comprising a diffraction enhancing element, wherein the diffraction enhancing element includes an antibody that is capable of binding to the analyte.

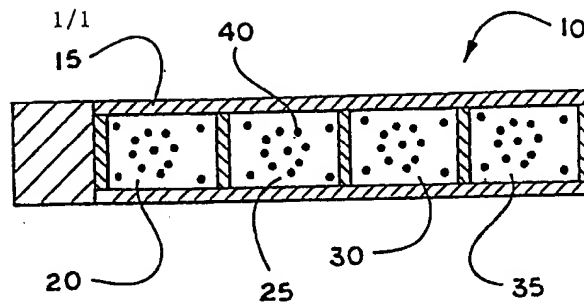


Fig. 1

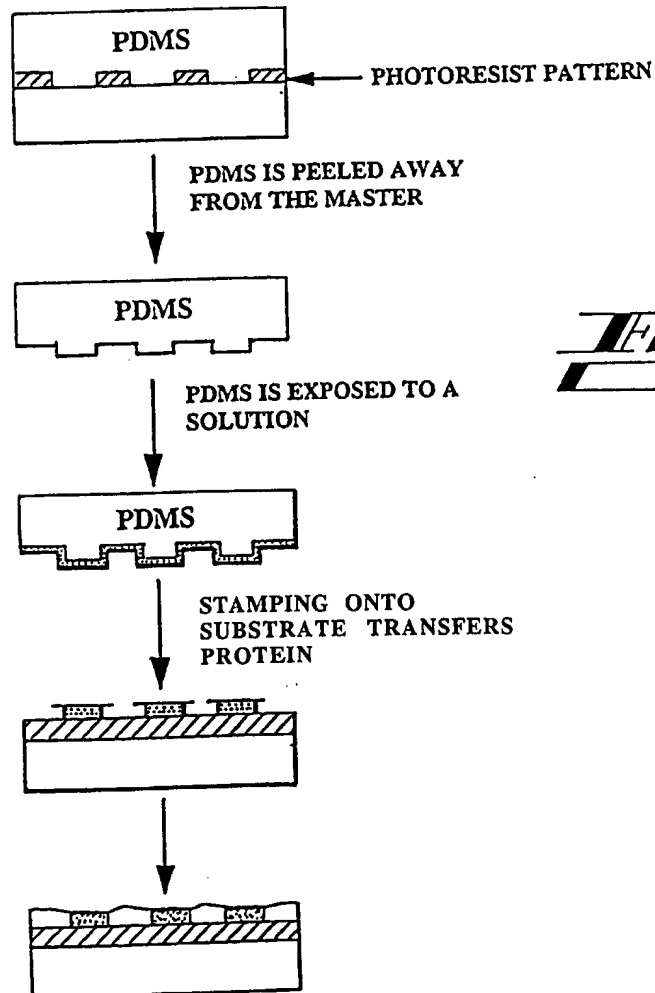


Fig. 2

INTERNATIONAL SEARCH REPORT

International Application No.
PCT/US 99/27727

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 G01N33/543 G01N21/47

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 G01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 98 27417 A (KIMBERLY CLARK CO) 25 June 1998 (1998-06-25) page 10, line 6 -page 11, line 5 claims ---	1-51
X	WO 96 09532 A (ABBOTT LAB) 28 March 1996 (1996-03-28) page 9, line 26-30 page 13, line 30 -page 14, line 7 page 27, line 19-27 --- -/--	1-6, 8, 13, 15, 20, 24, 26, 31-33, 36-45, 47

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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Date of the actual completion of the international search

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Muñoz, M

INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 99/27727

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	BLAWAS A S ET AL: "Protein patterning" BIOMATERIALS,GB,ELSEVIER SCIENCE PUBLISHERS BV., BARKING, vol. 19, no. 7-9, 5 April 1998 (1998-04-05), pages 595-609, XP004120826 ISSN: 0142-9612 the whole document ---	1-51
A	WO 98 21571 A (BINDER ANDRES ;CIBA GEIGY AG (CH); EHRAT MARKUS (CH); OROSZLAN PET) 22 May 1998 (1998-05-22) claims 1,2,10-13 -----	1-51

INTERNATIONAL SEARCH REPORT

Information on patent family members

Inter. Application No

PCT/US 99/27727

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 9827417 A	25-06-1998	US 5922550 A AU 5615698 A EP 0946865 A	13-07-1999 15-07-1998 06-10-1999
WO 9609532 A	28-03-1996	US 5599668 A AU 3636295 A CA 2197321 A EP 0783683 A JP 10506190 T US 5843651 A	04-02-1997 09-04-1996 28-03-1996 16-07-1997 16-06-1998 01-12-1998
WO 9821571 A	22-05-1998	AU 5479998 A	03-06-1998

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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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(21) International Application Number: PCT/US98/05400 (22) International Filing Date: 19 March 1998 (19.03.98) (30) Priority Data: 08/821,464 21 March 1997 (21.03.97) US (71) Applicant: KIMBERLY-CLARK WORLDWIDE, INC. [US/US]; 401 North Lake Street, Neenah, WI 54956 (US). (72) Inventors: EVERHART, Dennis, S.; 230 Hereford Road, Alpharetta, GA 30202 (US). KAYLOR, Rosann, M.; 7480 Williamsburg Drive, Cumming, GA 30131 (US). JONES, Mark, L.; 823 Lake Avenue, Atlanta, GA 30307 (US). (74) Agents: JOHNSON, James, Dean; Jones & Askew, 37th floor, 191 Peachtree Street, N.E., Atlanta, GA 30303 (US) et al.	(81) Designated States: AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, GM, GW, HU, ID, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG). Published <i>With international search report.</i> <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>	
(54) Title: GEL SENSORS AND METHODS OF USE THEREOF (57) Abstract The present invention comprises an optically diffracting sensing device whose diffraction pattern changes upon exposure to some stimuli. The diffraction pattern may be two or three dimensional, and in one embodiment the change in diffraction patterns is recognizable to the untrained eye. The device comprises one or more gels coated onto patterned, self-assembling monolayers of alkanethiolates, carboxylic acids, hydroxamic acids, and phosphonic acids printed onto a variety of substrates, including glass, silicon, aluminum oxide, and thermoplastic films metallized with gold, or with an alloy such as nickel/gold. The present invention also comprises the method of making this device, and the use of this device.		

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GEL SENSORS AND METHODS OF USE THEREOF

TECHNICAL FIELD

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The present invention is in the field of biosensors, and more specifically in the field of gel biosensors which rely on optical diffraction as the sensing mechanism.

BACKGROUND OF THE INVENTION

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Microcontact printing is a technique for forming patterns of organic monolayers with μm and submicron lateral dimensions. It offers experimental simplicity and flexibility in forming certain types of patterns. It relies on the remarkable ability of self-assembled monolayers of long-chain alkanethiolates to form on gold and other metals. These patterns can act as nanometer resists by protecting the supporting metal from corrosion by appropriately formulated etchants, or, can allow for the selective placement of fluids on hydrophilic regions of the pattern. Patterns of self-assembled monolayers having dimensions that can be less than $1\ \mu\text{m}$ are formed by using the alkanethiol as an "ink", and by printing them on the metal support using an elastomeric "stamp". The stamp is fabricated by molding a silicone elastomer using a master prepared by optical or X-ray microlithography or by other techniques.

Microcontact printing of patterned self-assembled monolayers brings to microfabrication a number of new capabilities. First, microcontact printing makes it possible to form patterns that are distinguished only by their constituent functional groups; this capability permits the control of surface properties such as interfacial free energies with great precision. Second, because microcontact printing relies on molecular self-assembly, it generates a system that is (at least locally) close to a thermodynamic minimum and is intrinsically defect-rejecting and self-healing. Simple procedures, with minimal protection against surface contamination by adsorbed materials or by particles, can lead to surprisingly low levels of defects in the final structures. The procedure can be conducted at atmospheric pressure, in an unprotected laboratory atmosphere. Thus, microcontact printing is especially useful in laboratories that do not have routine access to the equipment normally used in microfabrication, or for which the capital cost of equipment is a serious concern. Third, the patterned self-assembled monolayers can be designed to act as resists with a number of wet-chemical etchants.

Working with liquid etchants suffers from the disadvantages of handling solvents and disposing of wastes, but also enjoys substantial advantages: a high degree of control over contamination of surfaces; reduced damage to the substrate from energetic interactions with atoms or ions; the ability to manipulate complex and sensitive organic functionalities. Because the self-assembled monolayers are only 1 - 3 nm thick, there is little loss in edge definition due to the thickness of the resist; the major determinants of edge resolution seem to be the fidelity of the contact printing and the anisotropy of etching the underlying metal. In the current best cases, features of size 0.2 μm can be fabricated; edge resolution in systems showing this resolution in thickness is less than 50 nm.

Gels are cross-linked networks of polymers swollen with a liquid. Softness, elasticity, and the capacity to store a fluid make gels unique materials, and soft and gentle materials are beginning to replace some of the hard mechanical materials in various industries.

Due to the cross-linking, various properties of individual polymers become visible on a macroscopic scale. The polymer network changes its volume in response to a change in environment: temperature, solvent composition, mechanical strain, electric field, exposure to light, pH, salt concentration, etc.. Advances in Polymer Science, ed. K. Dusek, Vol. 109, p. v (Springer-Verlag New York 1993); S. Saito, pp. 207-232, *Id.*; M. Shibayama and T. Tanaka, pp. 1-62, *Id.*; Y. Osada, *et al.*, pp. 82-87, Scientific American (May 1993); Y. Osada and J. Gong, Prog. Polym. Sci., vol. 18, pp. 187-226 (Great Br. 1993); Irie, M., pp. 49-65 in Advances in Polymer Science, ed. K. Dusek, Vol. 110 (Springer-Verlag New York 1993); E. Kokufuta, pp. 157-77, *Id.*; T. Okano, pp. 179-197, *Id.*, all incorporated by reference.

Hydrophilic gels in aqueous solution have been the most widely studied, but almost any polymer can be cross linked to form a gel which will swell in a sufficiently good quality solvent. The three-dimensional network is stabilized by cross links which may be provided by covalent bonds, physical entanglements, crystallites, charge complexes, hydrogen bonding, van der Waal's or hydrophobic interactions. Gels have many technologically important roles in chemical separations, biomedical devices and absorbent products, to name a few areas. The properties that make gels useful include their sorption capacities, swelling kinetics, permeabilities to dissolved solutes, surface properties (*e.g.*, adhesiveness), mechanical characteristics, and optical properties. The single most important property of a gel is its swelling degree, since most of the properties are directly influenced by this. S.H. Gehrke, p. 85, in Advances in Polymer

Science, ed. K. Dusek, Vol. 110 (Springer-Verlag New York 1993).

"Responsive" polymer gels are materials whose properties, most notably their solvent-swollen volumes, change in response to specific environmental stimuli including temperature, pH, electric field, solvent quality, light intensity and wavelength, pressure, ionic strength, ion identity, and specific chemical triggers, like glucose. S. Saito, pp. 207-232; M. Shibayama and T. Tanaka, pp. 1-62. The property which often changes the most dramatically is the swollen volume. These changes may occur discontinuously at a specific stimulus level (a phase transition), or gradually over a range of stimulus values. All of these changes are reversible with no inherent limit in lifetime.

Gels have been employed as chemical sensing surfaces, for example, in conjunction with fiber-optic systems, or elaborate mechanical or electrode systems. These systems are often quite elaborate, and suffer either from lack of flexibility or expense, or both. For example, U.S. Patent No. 5,436,161 to J. Bergstrom, *et al.*, discloses a matrix coating for surface plasmon resonance detection, to be used with a rigid dielectric material, such as a glass plate.

The information-carrying capacity of light provides an elegant method for detecting and displaying information in a way that is readily interpreted by a human. Sensors that visibly change color in response to a surface antibody-antigen binding reaction are already commercially available. An example of such a device, based on thin film interference, is the group B streptococcal antigen detector made by Biostar™ [G. R. Bogart, *et al.* "Devices and methods for detection of an analyte based upon light interference. United States Patent No. 5,482,830, (Assignee: Biostar, Inc. Boulder, Colo.)]. Another example of a very simple optical-based sensor is where a Bragg reflector expands in the presence of water to change the reflected wavelength. The detection and display components

both device are integrated so that an electronic display (with associated power supply and processing circuit) is not needed.

However, that sort of detection device is suitable only for a narrow range of sensor applications. There is a need for a sensor technology platform that can be slightly modified to accommodate a wide range of stimuli and sensing conditions. There is a need, therefore, for a simple sensing system that takes full advantage of the responsive properties of gels, but which is flexible, easy to use, and preferably, disposable.

SUMMARY OF THE INVENTION

The present invention comprises an optically diffracting sensing device in which a diffraction pattern changes upon exposure to a predetermined stimuli. The diffraction pattern in the sensing device of the present invention may be two or three dimensional, and, in one embodiment, the change in diffraction patterns is recognizable to the untrained eye. The sensing device of the present invention comprises one or more gels coated onto patterned, self-assembling monolayers of alkanethiolates, carboxylic acids, hydroxamic acids, or phosphonic acids printed onto a variety of substrates, including, but not limited to, glass, silicon, aluminum oxide, and thermoplastic films metallized with gold, or with an alloy such as nickel/gold. The present invention also comprises the method of making this device, and the use of this device. In its desired embodiment, the sensing device of the present invention uses white light without any supporting detection or amplification systems.

Patterned self-assembling monolayers allow for the controlled placement of gel solutions thereon and which can contain a chemically reactive, indicator functionality. The gels suitable for use in the present invention can be produced by a variety of means, including solvent evaporation, radiation, or chemical cross-linking. When exposed to electromagnetic radiation, such as visible light, the sensing devices of the

present invention produce optical diffraction patterns which can change depending on the reaction of the gel with the stimulus of interest. The electromagnetic radiation can be in the visible spectrum, and can be either reflected from the substrate, or transmitted through the substrate. The stimulus to be detected can be any compound that reacts with the gel directly or with an indicator substance contained in the gel. (See, for example, Irie, M., pp. 49-65; Y. Osada and J. Gong) The present invention can be used to measure any stimuli to which a gel will respond, including, but not limited to, mechanical, temperature, electrical, and chemical stimuli.

These and other objects, features and advantages of the present invention will become apparent after a review of the following detailed description of the disclosed embodiments.

BRIEF DESCRIPTION OF THE FIGURES

Figure 1 is a schematic of contact printing of self-assembling monolayers, using a nickel/gold coated polymer substrate as an example. A polydimethylsiloxane (PDMS; silicone elastomer 184; Dow Corning Corp., Midland, MI) is polymerized on a silicone master containing a pre-determined pattern. The PDMS is peeled away from the master, and then exposed to a solution containing $\text{HS}(\text{CH}_2)_{15}\text{CH}_3$. The alkane-thiol coated stamp is then stamped onto the nickel/gold-coated substrate. Then, the surface of the substrate is exposed to a solution containing a different alkane thiol such as $\text{HS}(\text{CH}_2)_{11}\text{OH}$.

Figure 2 is a field emission secondary electron microscope image of 10 micron-diameter circles of hydrophilic self-assembling monolayers formed by printing of 16-mercaptohexadecanoic acid onto MYLAR® metallized with Ni/Au alloy, as described in Example 1.

Figure 3a is an optical photomicrograph at 300x magnification of 10 micron-diameter circles with center to center spacing of 15 microns of hydrophilic self-assembling

monolayers formed by printing of 16-mercaptohexadecanoic acid, as described in Example 1, below, and after exposure to a high surface energy, curable, optical adhesive. The adhesive is cured by ultraviolet light (UV) exposure.

5 Figure 3b is a photograph of the diffraction pattern formed by visible light shown through the self-assembling monolayer pattern described by Figure 3a.

10 Figure 4 is a field emission secondary electron micrograph image of 10 micron-diameter circles formed of self-assembled photocurable polymers on hydrophilic self-assembling monolayers, printed as described in Example 1.

15 Figure 5 is an atomic force microscopy image of evaporated gold on MYLAR®, purchased from Courtaulds Performance Films (Canoga Park, CA). The average roughness of the gold layer is 3-4 nanometers, with maximum roughness of 9 nanometers.

 Figure 6, in the left hand vertical column shows printed arrays, on in the right-hand column shows the resulting diffraction patterns.

20 Figure 7, as described in Example 14, at the left hand side is an illustration of a printed hologram, and the corresponding smiling face image it produces. The right hand side of Figure 7 are two enlarged views, side plan and top plan, of the circles of self-assembling monolayers which all together make up the printed hologram.

25 Figure 8, shows a graph of a temperature induced volume transition in a gel, where the ordinate gives the temperature in degrees Celsius, and the abscissa gives the ratio of resultant volume to original volume. The right hand side of Figure 8 is a schematization of a volume change of a gel due to factors such as solvent composition, temperature, ions, pH, light, and electric field.

30 Figure 9, on the left hand side, shows a hologram diffraction pattern printed on high resolution printer film, as described in Example 13. The right hand side shows a

35

responsive gel atop the same diffraction pattern on metallized Mylar®. The intermediate steps are not shown.

Figure 10 is an illustration of a micro-bump array, as described in Example 7, where the bump center to center spacing is 15 microns.

Figure 11 is an illustration of a side view of a typical micro-bump array, as described below and in Example 7, showing that the diffraction angle θ and the number of visible diffraction orders are dependent on the incident wavelength λ , and the bump center-to-center spacing Λ .

Figure 12 illustrates a typical far field diffraction pattern, as described below, for micro-bump array with a 15 micron period (left-hand side) and for one with a 3 micron period (right-hand side).

Figure 13, as described below, graphically illustrates the transmitted far field electric field magnitude $|T(u)|$ as periodic, because the light wave front is periodically perturbed (delayed) as it travels through the higher refractive index micro-bumps, where each order's electric field (a_0, a_1, a_2, \dots) is related to the perturbed wave front's Fourier series coefficient.

Figure 14, as described below, illustrates a calculated half-plane, on-axis diffraction pattern irradiance for two different micro-bump arrays with refractive index = 1.5, period $\Lambda = 15$ micrometers, bump width = 10 micrometers, bump height = 1.5 micrometers (large dots), bump height = 0.9 micrometers (small dots). The abscissa is in degrees, and the ordinate is measured in power % of total transmittal of irradiance.

Figure 15, as described in Example 4, illustrates an experimental setup for quantifying the diffraction spectra from a diffraction array.

Figure 16, as described in Example 4, illustrates a diffraction image from poly-N-isopropylacrylamide micro-

bump sensor array, where the zero-order beam was blocked to eliminate CCD saturation.

Figure 17, as described in Example 4, is a graph of the first-order diffraction irradiance as a function of the micro-bump height for a bump refractive index of 1.5, base width of 10 micrometers, and a period of 15 micrometers. The oscillating irradiance curve limits the useful bump height to approximately 0.3 micrometers for this geometry. The abscissa measure bump height in microns, and the ordinate measures irradiance in power % of total transmittal of irradiance.

Figure 18, as described in Example 5, illustrates a holographic recording setup, with an object plane, a film plane, a signal beam, and a reference beam, with a smiling face as the holographic image.

Figure 19, as described in Example 7, illustrates the twin images arising from the amplitude hologram, the virtual image on the left-hand side of Figure 19, and the real image on the right-hand side, with the viewer positioned at the lower right-hand corner.

Figure 20, as described in Example 7, illustrates conjugate beam haze. The in-focus image is the real image, the large dot is the transmitted beam, and the haze is virtual image. The projected image on the left is illuminated through hologram center. Image on right is illuminated slightly off-center to illustrate the symmetric conjugate image placement.

Figure 21, as described in Example 9, shows a recognizable object on the left-hand side, and its associated hologram on the right-hand side.

Figure 22, as described in Example 9, shows a 3600 dots per inch (dpi) test page printout with 20 test holograms on one page.

Figure 23, as described in Example 13, shows several pattern transfer steps from the printer output on high-resolution printer film (A) to the photoresist master on gold film (B) to the elastomer stamp (C) to the responsive gel on metallized Mylar® (D). The small line in (C) is 30 microns long.

Figure 24, as described in Example 14, illustrates a method of projecting the real image onto a screen, showing the incident light, the film plane, and the image plane.

DETAILED DESCRIPTION

The present invention comprises an optically diffracting sensing device in which the diffraction pattern changes upon exposure to predetermined stimuli. The diffraction pattern may be two or three dimensional and, in one embodiment, the change in diffraction patterns is recognizable to the unaided eye. The sensing device of the present invention further comprises one or more gels coated onto patterned, self-assembling monolayers of, for example, alkanethiolates, carboxylic acids, hydroxamic acids, or phosphonic acids printed onto a variety of substrates including, but not limited to, glass, silicon, aluminum oxide, and thermoplastic films metallized with gold, or with an alloy such as nickel/gold. Other substrates that can be used according to the present invention include, but are not limited to, CrOx, CuOx, AgOx, platinum, and lead. The gels used according to the present invention are capable of responding to the presence or change in stimuli. Usually the response is a change in volume, shape, color or a change in refractive index. The stimuli can be a chemical compound or a physical parameter such as environment: temperature, solvent composition, mechanical strain, electric field, exposure to light, pH, salt concentration, solvent quality, light intensity and wavelength, pressure, ionic strength, ion identity, and specific chemical triggers, e.g.,

glucose. The present invention also comprises the method of making the sensing device and the use of this device.

5 Patterned self-assembling monolayers allow for the controlled placement of the gel thereon. The term "patterned self-assembling monolayers thereon" as used herein means the self-assembling monolayers in any pattern on the metallized polymer films including a solid pattern. The optical sensing devices of the present invention produce optical diffraction patterns which differ depending on the reaction of the self-assembling monolayer and the gel with the stimulus of interest. 10 The electromagnetic radiation that is diffracted is preferably in the visible spectrum, and can be either reflected from the substrate or transmitted through the substrate. The stimulus to be measured by the sensing device of the present invention can be any substance or physical parameter that interacts with the gel or with an analyte in the gel. It is contemplated as part of the invention that the gel can contain an analyte that will react with a stimulus thereby causing the gel to change in volume, shape, color or refractive index. Thus, it can be the gel 20 interacting directly with the stimulus or the gel can contain an analyte that reacts with the stimulus.

Microcontact printing is a technique for forming patterns of organic monolayers with micron or submicron lateral dimensions. It offers experimental simplicity and 25 flexibility in forming certain types of patterns. It relies on the remarkable ability of self-assembled monolayers of long-chain alkanethiolates to form on gold and other metals. These patterns can act as nanometer resists by protecting the supporting substrate from corrosion by appropriately formulated etchants, or, can allow for the selective placement of fluids on hydrophilic regions of the pattern. Patterns of self-assembled monolayers having dimensions that can be less than 1 μm are formed by using the alkanethiol as an "ink", and by printing them on the metal support using an elastomeric 30 "stamp". The stamp is fabricated by molding a silicone elas-

tomers using a master prepared by optical or X-ray microlithography or by other techniques.

Microcontact printing of patterned self-assembled monolayers brings to microfabrication a number of new capabilities. First, microcontact printing makes it possible to form patterns that are distinguished only by their constituent functional groups; this capability permits the control of surface properties such as interfacial free energies with great precision. Second, because micro-contact printing relies on molecular self-assembly, it generates a system that is (at least locally) close to a thermodynamic minimum and is intrinsically defect-rejecting and self-healing. Simple procedures, with minimal protection against surface contamination by adsorbed materials or by particles, can lead to surprisingly low levels of defects in the final structures. The procedure can be conducted at atmospheric pressure, in an unprotected laboratory atmosphere. Thus, microcontact printing is especially useful in laboratories that do not have routine access to the equipment normally used in microfabrication, or for which the capital cost of equipment is a serious concern. Third, the patterned self-assembled monolayers can be designed to act as resists with a number of wet-chemical etchants.

Because the self-assembled monolayers are only 1 - 3 nm thick, there is little loss in edge definition due to the thickness of the resist; the major determinants of edge resolution seem to be the fidelity of the contact printing and the anisotropy of etching the underlying metal. In the current best cases, features of size 0.2 μm can be fabricated; edge resolution in systems showing this resolution in thickness is less than 50 nm.

Gels, as used in the present invention, have both liquid-like and solid-like properties. The liquid-like properties result from the fact that the major constituent of gels is usually a liquid, *e.g.*, water. For example, a jelly consists of

approximately 97% water and 3% gelatin. On the other hand, a gel can retain its shape since it has a shear modulus which becomes apparent when the gel is deformed. The modulus is due to the cross-linking of the polymers in the form of a network. These aspects of a gel represent the solid nature of gels. In addition to these liquid- and solid-like aspects, a gel can change its state drastically, similar to the way a gas changes its volume more than a thousand fold. Two states of gels; the collapsed and swollen states, correspond to the liquid and the gas states of fluids respectively. Figure 8 shows a graph of a temperature induced volume transition in a gel, where the ordinate gives the temperature in degrees Celsius, and the abscissa gives the ratio of resultant volume to original volume. As can be seen by the graph, a sharp volume transition occurs for this gel between 34 and 36 degrees Celsius. The right hand side of Figure 8 is a schematization of a volume change of a gel due to factors such as solvent composition, temperature, ions, pH, light, and electric field.

A gel can be viewed as a container of solvent made of a three dimensional mesh. In a dried state, a gel is a solid material. However, a gel swells until it reaches the swelling equilibrium when a solvent is added. The solvent molecules are kept in the three dimensional mesh and the combination of the mesh and the solvent molecules creates a "world" having characteristic properties. This world can be either isolated from (isochore) or linked to (isobar) its surrounding world by changing the population, *i.e.*, the solvent molecules.

A gel can be a "single polymer molecule". The term "single polymer molecule" means that all the monomer units in a one piece of gel are connected to each other and form one big molecule on a macroscopic scale. Because of this nature, a gel is a macroscopic representation of single polymer behavior.

Many kinds of external stimuli, such as, temperature, pH, photons, ions, electric current (field), etc., can control the

volume of the gel. Particularly, in the case of volume phase transition, an enormous change in volume can be induced by an infinitesimal change of one of the these stimuli, and this is of great importance in the present invention, as an actuator,
5 sensor, switching device and so on. See Figure 8. These volume phase transitions can be induced by van der Waals, hydrophobic, hydrogen bonding, electrostatic, and charge-transfer interactions.

In the present invention, the gel is coupled to the self-assembling monolayer printed as described above. Depending
10 upon the property to be sensed, the gel includes, but is not limited to, a polysaccharide such as agarose, dextran, carageenan, alginic acid, starch, cellulose, deionized gelatin, and derivatives of these such as carboxymethyl derivatives. The gel may also be a water-swellaable organic polymer such as
15 polyvinyl alcohol, polyacrylic acid, polyacrylamide, or polyethylene glycol. Aqueous gels are also known in the art as "hydrogels", and "hydrophilic polymers". They may be copolymers or homopolymers. Suitable copolymers may
20 either be regular copolymers containing substantially no other material in their matrices, or they may be copolymers which contain monomers such as styrene and vinyl acetate, for example. Examples of suitable copolymers which may or may not contain monomers include, but are not limited to, N-vinyl
25 pyrrolidone and glycidyl methacrylate.

Homopolymers include those that are slightly cross-linked, such as hydroxyethyl methacrylate. Suitable copolymers with or without monomers and homopolymers may also be polymerized from the following non-limiting list
30 of monomers: hydroxyalkyl acrylates and hydroxyalkyl methacrylates, for example, hydroxyethyl acrylate, hydroxypropyl acrylate, and hydroxybutyl methacrylate; epoxy acrylates and epoxy methacrylates, such as, glycidyl methacrylate; amino alkyl acrylates and amino alkyl
35 methacrylates; N-vinyl compounds, such as, for example, N-

vinyl pyrrolidone, N-vinyl carbazole, N-vinyl acetamide, and N-vinyl succinimide; amino styrenes; polyvinyl alcohols and polyvinyl amines; polyacrylamides such as N-isopropyl acrylamide and various substituted polyacrylamides; vinyl pyridine; vinyl sulfonate and polyvinyl sulfate; vinylene carbonate; vinyl acetic acid, and vinyl crotonic acid; allyl amine and allyl alcohol; and vinyl glycidyl ethers.

Polymer gels swollen in nonvolatile organic solvents are known as organogels. A few examples of organogels are systems based on aluminum stearate, oleate, or naphthenate. These polymers form non-aqueous gels on cooling with hydrocarbons. Electroconductive organogels have also been prepared from 3-alkyl thiophenes using FeCl₃ as a catalyst. These polymer gels exhibit drastic volume changes when the solvent composition of ethanol-chloroform was changed. The absorption spectrum also changes in association with the volume change and temperature. Another type of electroconductive gel consists of an electrodonating polymeric network and a low molecular weight acceptor subsequently doped to the gel. Thus, a 7,7,8,8-tetracyanoquinodimethane (TCNQ) was doped as an electron acceptor into a cross linked polymeric donor: poly [N-[3-(dimethylamino)propyl]acrylamide] (PDMA PAA) in dimethylformamide (DMF). When TCNQ was doped, a significant swelling and coloration due to the formation of a charge-transfer (CT) complex occurred.

The gel can be derivatized to contain hydroxyl, carboxyl, amino, aldehyde, carbonyl, epoxy, or vinyl groups for immobilizing a desired ligand, and optionally, a biospecific ligand bound via said groups. Many examples of suitable gels may be found in *Hydrogels in Medicine and Pharmacy*, vols. I-III, ed. Peppas, N.A. (CRC Press 1986-7), and *Advances in Polymer Science*, ed. K. Dusek, vols. 109, 110 (Springer-Verlag New York 1993), and references cited therein, all incorporated herein by reference.

Examples of derivatization of gels may be found in U.S. Patent no. 5,436,161 to J. Bergström, which is incorporated herein by reference. In one embodiment of the present invention, the desired ligand might be an antibody, a T or B cell receptor, an epitope, or a fragment of any of the foregoing including, but not limited to, molecules, such as proteins, glycoproteins, metal salts, ions, and the like. The gel may also include neurotransmitters, hormones, growth factors, cytokines, monokines, lymphokines, nutrients, enzymes, and receptors. Also included are structured elements such as macromolecular structures, organelles and cells, including, but not limited to, cells of ectodermal, mesodermal, and endodermal origin such as stem cells, blood cells, neural cells, immune cells, and gastrointestinal cells, and also microorganisms, such as fungi, viruses, bacteria and protozoa. Many of these gel encapsulated cells can produce a volume changing stimulus when exposed to specific analytes.

In one embodiment of the present invention, a hydrazide function is created in the dextran matrix for binding ligands containing aldehyde groups, for example antibodies, in which the carbohydrate chain has been oxidized so that it then contains an aldehyde function. In this instance, the dextran matrix is initially modified with carboxymethyl groups which are partly reacted to form hydrazide groups. With this activated matrix at least two important advantages are obtained: (1) This matrix contains unreacted carboxyl groups which in low ionic strength conditions will act as ion exchangers, and by electrostatic interaction the ligand which is to be immobilized is connected to the dextran matrix; (2) This matrix will very efficiently bind the ligand thus concentrated at the surface, viz. by condensation of ligand aldehyde groups with the hydrazide function of the matrix.

According to another embodiment of the present invention, a part of the carboxyl groups in carboxymethyl-modified dextran are modified so as to give

reactive ester functions, *e.g.*, by treatment with an aqueous solution of N-hydroxysuccinimide and N-(3-dimethyl-aminopropyl)-N'-ethylcarbodiimide hydrochloride. In the same way as in the example described above, the residual charges, *i.e.*, unreacted carboxyl groups, will contribute to effecting a concentration of ligands on the surface. Ligands containing amine groups such as, for example, proteins and peptides, may then be coupled to the dextran matrix by covalent bonds.

According to an alternative procedure, the aforesaid reactive ester is utilized for reaction with a disulfide-containing compound such as, for instance, 2- (2-pyridinyldithio) ethanamine: in this manner a matrix is obtained which contains disulfide groups, and these can be employed for coupling thiol-containing ligands such as, for example, reduced F(ab) fragments of immunoglobulins (*see* Brocklehurst, K., *et al.*, J. Biochem., vol. 133, p. 573, *et seq.* (1973), incorporated herein by reference). After cleavage of the disulfide bonds, for instance, by reduction or thioldisulfide exchange, the thiol modified surface formed can be used for coupling of a disulfide-containing ligand such as, for instance, N- succinimidyl 3-(2-pyridinyldithio) propionate (SPDP) modified proteins.

The advantage of this procedure is that the ligands via, for example, a reduction step can be cleaved off to give a sensing surface with reactive thiols. This thiol-modified surface can, in an analogous procedure, be used for renewed covalent coupling of thiol- or disulfide-containing ligands. In this way the capability of chemical regeneration of the sensing surface can be obtained, which can be used for general utilization of the same surface for couplings of several different ligands. The procedure can also be used when, for example, a biological interaction is studied, and this interaction cannot be broken while retaining biological activity of the immobilized ligand.

One important aspect of the present invention is that one or more of the layers forming the sensing surface to be used in a given analysis can be synthesized and/or functionalized *in situ* by adding the appropriate reagents to the surface in a flow-through cell in a biosensor system.

In summary, there are a multitude of ligands that can be employed for the detection of biomolecules by means of interacting therewith. It will be readily evident that ion exchanging groups, metal chelating groups and various types of receptors for biological molecules - known from conventional liquid chromatographic procedures - may be employed for the construction of systems which are suitable for selection purposes, even in complex measuring systems.

Metallo-organic materials, such as metallo-phthalocyanine, may also be included in the gel. Other substances, such as surfactants, inorganic salts, *e.g.*, NaBr, KBr, NaCl, KCl, NaI, and KI, polar organic additives, such as methanol and glycerol, tetra-alkylammonium bromides, and crown ethers, *e.g.*, benzo[18]crown-6, may be added to the gel to affect its swelling characteristics. Saito, Konno & Inomata; Irie. Light sensitive compounds, such as azobenzene chromophores, can be added to the gel to affect its characteristics.

The gel used in the present invention may also be made in a gradient arrangement, as set forth in "Molecular Gradients of Substituted Alkanethiols on Gold: Preparation and Characterization", by Bo Liedberg and Pentti Tengvall, published in *Langmuir*, Vol. 11, No. 10, 1995, pp. 3821-3827.

When the substrate with the gel atop the self-assembling monolayers is exposed to a stimulus or analyte that is capable of reacting with or affecting the gel, the sensing device produces optical diffraction patterns which change from the original optical diffraction pattern, depending on the reaction of the gel on the self-assembling monolayer with the stimulus of interest. It is to be understood that more than one self-

assembling monolayer can be printed on a substrate thereby allowing one to associate the gel with one self-assembling monolayer and not with the second self-assembling monolayer.

5 Self-assembled monolayers of organic compounds on inorganic or metal surfaces are becoming increasingly important in many areas of materials science. Although there are many different systems of self-assembling monolayers based on different organic components and supports, desired systems are those of alkanethiolates, $\text{HS}(\text{CH}_2)_n\text{R}$. Typically, a
10 gold film, 5 to 2000 nm thick, is supported on a titanium-primed Si/SiO₂ wafer or glass sheet. The titanium serves as an adhesion promoter between gold and the support. The alkanethiols chemisorb on the gold surface from a solution in which the gold film is immersed, and form adsorbed alkanethiolates with loss of hydrogen. Adsorption can also
15 occur from the vapor. Self-assembling monolayers formed on gold from long-chain alkanethiolates of structure $\text{X}(\text{CH}_2)_n\text{Y}-(\text{CH}_2)_m\text{S}$ are highly ordered and can be considered as crystalline or quasi-crystalline molecular arrays. A wide
20 variety of organic functional groups (X,Y) can be incorporated into the surface or interior of the monolayer.

Self-assembling monolayers can therefore be tailored to provide a wide variety of material properties: wettability and protection against corrosion by chemical etchants are
25 especially relevant to μCP . In one embodiment of the present invention, there are two or more self-assembling monolayers with different chemical properties. In another embodiment of the present invention, a first self-assembling monolayer is hydrophobic, and a second self-assembling monolayer is
30 hydrophilic.

Figure 1 outlines the procedure used for microcontact printing onto a substrate. An elastomeric stamp is used to transfer by contact alkanethiol "ink" to a surface coated with a metal alloy. In a desired embodiment, the alloy surface is
35 predominantly gold. Preferred alloys are those such as

nickel/gold, which are known to show an enrichment in the surface concentration of gold relative to its bulk concentration. Prediction of surface segregation of one metal of an alloy is described in M.P. Seah, "Quantitative Prediction of Surface Segregation," Journal of Catalysis, vol. 57, pp. 450-457 (1979), and J.J. Burton, *et al.*, "Prediction of Segregation to Alloy Surfaces from Bulk Phase Diagrams," Physical Review Letters, vol. 37, No. 21, pp. 1433-1436 (Nov. 22, 1976), both incorporated herein by reference. In one embodiment of the invention, the metal alloy has surface enrichment of a metal reacting with the self-assembling monolayer. If the stamp is patterned, a patterned self-assembling monolayer forms. The stamp is fabricated by casting polydimethylsiloxane (PDMS) on a master having the desired pattern. Masters are prepared using standard photolithographic techniques, or constructed from existing materials having microscale surface features. These methods are disclosed in U.S. Patent No. 5,512,131 and copending U.S. Patent Application Serial No. 08/707,456 entitled, "Method of Contact Printing on Metal Alloy Coated Polymer Films," and the U.S. patent application entitled, "Method of Contact Printing on Gold Coated Films," filed December 18, 1996, all of which are incorporated herein by reference.

In a typical procedure, a photolithographically produced master is placed in a glass or plastic Petri dish, and a 10:1 ratio (w:w or v:v) mixture of SYLGARD silicone elastomer 184 and SYLGARD silicone elastomer 184 curing agent (Dow Corning Corporation) is poured over it. The elastomer is allowed to sit for approximately 30 minutes at room temperature and pressure to degas, then cured for 1 to 4 hours at 60°C, and gently peeled from the master. "Inking" of the elastomeric stamp is accomplished by exposing the stamp to a 0.1 to 10 mM solution of alkanethiol in anhydrous ethanol, either by pouring the solution over the surface of the stamp, or by rubbing the stamp gently with a Q-tip that has been

5 saturated with the inking solution. The stamp is allowed to dry until no liquid is visible by eye on the surface of the stamp (typically about 60 seconds), either under ambient conditions, or by exposure to a stream of nitrogen gas. Following inking, the stamp is applied to a metal alloy, *e.g.*, nickel/gold surface. Very light hand pressure is used to aid in complete contact between the stamp and the surface. The stamp is then gently peeled from the surface. Following removal of the stamp, the surface is washed of excess thiol and the patterned metal alloy surface can be subjected to chemical etchants (see below) that selectively remove underivatized areas of the metal alloy surface, and if desired, the underlying support(s). Alternatively, further derivatization of unstamped areas can be accomplished, either by using a second stamp, or by washing the entire surface with a different alkanethiol.

10 The elastomeric character of the stamp is essential to the success of the process. Polydimethylsiloxane (PDMS), when cured, is sufficiently elastomeric to allow good conformal contact of the stamp and the surface, even for surfaces with significant relief; this contact is essential for efficient contact transfer of the alkanethiol "ink" to the alloy-coated film. The elastomeric properties of PDMS are also important when the stamp is removed from the master. If the stamp is rigid (as is the master) it is difficult to separate the stamp and master after curing without damaging one of the two substrates. PDMS is also sufficiently rigid to retain its shape, even for features with sub-micron dimensions. Patterns with lines as small as 200 nm in width have been generated. The surface of PDMS has a low interfacial free energy ($\gamma = 22.1$ dynes/cm), and the stamp does not adhere to the metal alloy coated film. The stamp is durable. The same stamp has been used up to 100 times over a period of several months without significant degradation in performance. The polymeric nature of PDMS also plays a critical role in the inking procedure by enabling the stamp to absorb the alkanethiol ink by swelling.

Microcontact printing on metal alloy surfaces can be conducted with a variety of alkanethiol "inks". Alkanethiols that do not undergo reactive spreading (after application to the metal alloy film) are required for formation of small features with high resolution. For stamping in air, one can use autophobic alkanethiols such as hexadecanethiol. Microcontact printing of other non-autophobic alkanethiols, for example, $\text{HS}(\text{CH}_2)_{15}\text{COOH}$, can be conducted by stamping under a liquid such as water. Patterned self-assembling monolayers of alkanethiols on metal alloy provide excellent resist character with a number of wet-chemical etchants.

In one embodiment of the present invention, the self-assembling monolayer is formed of a carboxy-terminated alkane thiol stamped with a patterned elastomeric stamp onto a nickel/gold-surfaced thermoplastic film such as MYLAR[®]. The stamp is inked with a solution of alkanethiol in ethanol, dried, and brought into contact with a surface of nickel/gold. The alkanethiol is transferred to the surface only at those regions where the stamp contacts the surface, producing a pattern of self-assembling monolayer which is defined by the pattern of the stamp. Optionally, areas of unmodified nickel/gold surface next to the stamped areas can be rendered hydrophobic by reaction with a methyl-terminated alkane thiol. The film is then contacted with a solution capable of forming a gel, *e.g.*, an aqueous solution of N-isopropylacryamide: for example, to coat the film it may be drawn through a two phase system of water and toluene. The polymer then assembles onto the patterned, hydrophilic SAM, forming the sensing device of the present invention.

A desirable embodiment of the present invention is a thermoplastic film substrate upon which the SAM with the gel is placed. Any thermoplastic film upon which a metal substrate can be deposited is suitable for the present invention. These include, but are not limited to polymers such as: polyethylene-terephthalate (MYLAR[®]), acrylonitrile-

butadiene-styrene, acrylonitrile-methyl acrylate copolymer, cellophane, cellulosic polymers such as ethyl cellulose, cellulose acetate, cellulose acetate butyrate, cellulose propionate, cellulose triacetate, cellulose triacetate, 5 polyethylene, polyethylene - vinyl acetate copolymers, ionomers (ethylene polymers) polyethylene-nylon copolymers, polypropylene, methyl pentene polymers, polyvinyl fluoride, and aromatic polysulfones. Preferably, the plastic film has an optical transparency of greater than 80%. Other suitable 10 thermoplastics and suppliers may be found, for example, in reference works such as the *Modern Plastics Encyclopedia* (McGraw-Hill Publishing Co., New York 1923-1996), incorporated herein by reference.

In one embodiment of the invention, the thermoplastic 15 film with the metal coating thereon has an optical transparency of between approximately 5% and 95%. A more desired optical transparency for the thermoplastic film used in the present invention is between approximately 20% and 80%. In a desired embodiment of the present invention, the 20 thermoplastic film has at least an approximately 80% optical transparency, and the thickness of the metal coating is such as to maintain an optical transparency greater than about 20%, so that diffraction patterns can be produced by either reflected or transmitted light. This corresponds to a metal coating 25 thickness of about 20 nm. However, in other embodiments of the invention, the gold thickness may be between approximately 1 nm and 1000 nm.

The preferred metal alloy for deposition on the film is gold and another metal. However, alloys of silver, aluminum, 30 copper, iron, zirconium, platinum, nickel may also be used. Preferred metals are ones that do not form oxides, and thus assist in the formation of more predictable self-assembling monolayers. Alloys such as Ni/Au, Pt/Au, and Cu/Au, which show surface enrichments of Au, are suitable.

In principle, any surface with corrugations of appropriate size could be used as masters. The process of microcontact printing starts with an appropriate relief structure, from which an elastomeric stamp is cast. This 'master' template may be generated photolithographically, or by other procedures, such as commercially available diffraction gratings. In one embodiment, the stamp may be made from polydimethylsiloxane.

In one embodiment of the present invention, the self-assembling monolayer has the following general formula:



X is reactive with metal or metal oxide. For example, X may be asymmetrical or symmetrical disulfide (-R'SSR, -RSSR), sulfide (-R'SR, -RSR), diselenide (-R'Se-SeR), selenide (-R'SeR, -RSeR), thiol (-SH), nitrile (-CN), isonitrile, nitro (-NO₂), selenol (-SeH), trivalent phosphorous compounds, isothiocyanate, xanthate, thiocarbamate, phosphine, thioacid or dithioacid, carboxylic acids, hydroxylic acids, and hydroxamic acids.

R and R' are hydrocarbon chains which may optionally be interrupted by hetero atoms and which are preferably non-branched for the sake of optimum dense packing. At room temperature, R is greater than or equal to seven carbon atoms in length, in order to overcome natural randomizing of the self-assembling monolayer. At colder temperatures, R may be shorter. In a preferred embodiment, R is -(CH₂)_n- where n is between 10 and 12, inclusive. The carbon chain may optionally be perfluorinated. See Regen, S.L., *et al.*, (1986), J. Am. Chem. Soc., vol. 108, pp. 6094-5, and Ringsdorf, H., *et al.*, Angew. Chem. Int. Ed., Engl., vol. 27, pp. 113-158 (1988), both incorporated herein by reference.

Y may also have any surface property of interest. For example, Y could be any among the great number of groups

used for immobilization in liquid chromatography techniques, such as hydroxy, carboxyl, amino, aldehyde, hydrazide, carbonyl, epoxy, or vinyl groups. Examples of sensing layer materials are set forth in "Patterning Self-Assembled Monolayers Using Microcontact Printing: A New Technology for Biosensors?," by Milan Mrksich and George M. Whitesides, published in TIBTECH, June, 1995 (Vol. 13), pp. 228-235; and U.S. Patent No. 5,436,161 to J. Bergstrom, *et al.*, hereby incorporated by reference.

In one embodiment of the invention, the gel is coupled to Y. For example, both the gel and the SAM may be hydrophilic.

Self-assembling monolayers of alkyl phosphonic, hydroxamic, and carboxylic acids may also be useful for the methods and compositions of the present invention. Since alkanethiols do not adsorb to the surfaces of many metal oxides, carboxylic acids, phosphonic acids, and hydroxamic acids may be preferred for X for those metal oxides. See J. P. Folkers, G.M. Whitesides, *et al.*, *Langmuir*, 1995, vol. 11, pp. 813-824.

R may also be of the form $(CH_2)_a-Z-(CH_2)_b$, where $a \geq 0$, $b \geq 7$, and Z is any chemical functionality or compound of interest, such as sulfones, urea, lactam, etc.

The stamp may be applied in air, or under a fluid such as water to prevent excess diffusion of the alkanethiol. For large-scale or continuous printing processes, it is most desirable to print in air, since shorter contact times are desirable for those processes.

In one embodiment of the present invention, the pattern is formed on the metallized thermoplastic polymer with the self-assembling monolayer. In another embodiment of the present invention, the relief of the pattern is formed with the self-assembling monolayer. After the stamping process, the metallized areas on the plastic may optionally be passivated,

for example, with a methyl-terminated self-assembling monolayer such as hexadecylmercaptan.

5 The appearance, or disappearance, of a holographic image can be used to indicate the presence of a stimulus in the local environment, thus, holograms can be used to simplify such a device and to present the display information to a consumer in a user-friendly fashion. A computer algorithm is used to calculate and generate diffraction hologram patterns of pre-defined objects.. Transfer of the computer-generated
10 pattern to a responsive gel on metallized Mylar® was accomplished via a simple printing process.

Applications for optical holography include digital data storage, microscopy, spectroscopy pattern recognition, and displays. Handbook of Optical Holography (H. J. Caulfield,
15 editor), New York. Academic Press 1979, incorporated herein by reference. Many of these applications rely on traditional holographic methods which are not well suited to the requirements of a sensor device.

20 The present invention allows one to computer-generate a complex holographic pattern of a specified object, print it on high resolution film, and convert the patterned elastomeric stamp for final transfer to a substrate upon which a selectively responsive material can be self-assembled to form the light-diffracting hologram pixels.

25 The ability to easily view a holographic image with the unaided eye involves consideration of a number of variables including (1) lighting conditions, (2) the refractive index modulation amplitude in the holographic material, (3) the hologram information content, (4) the type of hologram
30 (volume phase, amplitude, transmission, reflection, etc.), (5) the thickness of the active diffraction layer, and (6) the hologram's activated pixel population. The use of a hologram for a sensor application requires precise receptor patterning, micro-reactor site activation by the stimulant, and a

corresponding local change in light absorption or refractive index.

In the present invention, sensor information extracted from the light pattern diffracting from a regular array of micro-bumps is quantified and related to physical changes (size, shape, and refractive index) at the micro-bump level.

According to the present invention, the hologram pattern is fabricated for sensor applications. The method according to the present invention is compatible with continuous-print processes and it involves the following steps: (a) computation of the printed hologram pattern based on a preselected visual image to display, (b) formatting and printing the pattern on high-resolution transparency film, (c) photolithographic conversion of the pattern from gray-scale to surface-relief, (d) conversion of the surface-relief pattern to an elastomeric stamp, (e) stamp printing hydrophilic/hydrophobic mono-layer regions on a metallized MYLAR® substrate, and finally, (f) assembling analyte-responsive material on regions defined in the stamp printing process.

A special light source, such as a laser pointer or a white-light point source, is desirable for viewing the printed holographic image. Each location in the printed hologram contains information about a perspective view of the whole image. Therefore, when a laser pointer is used to project the image onto a screen, the beam only needs to propagate through a small section of the hologram to reconstruct the whole image. Alignment is not critical.

According to the present invention, a hologram pattern printed on a two-dimensional (surface) substrate represents only the real part of the complex light propagation information. Therefore, both a real and a symmetric conjugate (virtual) image are reconstructed during viewing. The "dual" image formation lends an added degree of flexibility but it also

effectively reduces the unobscured, viewable image space by half.

5 The size, relative position, and information content of the holographic image are all limited by the resolution capabilities of the printer. A 3600 dot per inch (dpi) printout limits the image full fan angle to just over 5 degrees (the farther away from the film, the larger the image can be). Furthermore, to avoid overlap with the twin image, the effective image fan angle for a hologram generated with a maximum 3600 dpi resolution is reduced to 2.5 degrees. 10 Methods exist whereby a 10 times photolithographic reduction of the hologram pattern can be used to increase the effective density of the pixels to 36,000 dpi, thereby, increasing the effective (unobscured) image fan angle to 32 degrees. If 15 higher resolution is necessary conventional photo or electron beam lithographic processes can be used.

The diffracted image can be optimized for the specific method of indication. For example, an image as simple as a single, off-axis, projected dot may be ideal when using a photo-diode for detection, but a complex symbol image such as a skull and crossbones may be better suited for detection by the human eye. Each type of image can be specified; however, the hologram computation time increases linearly with each added image pixel. 20

25 A diffraction based sensor according to the present invention utilizes the interaction of light with a sensing medium to alter the transmitted or reflected diffraction pattern. Figure 10 shows a periodic structure consisting of polymer micro-bumps patterned in an array. A change in the array's micro-bump size, shape, color, optical density, or 30 refractive index may be triggered by the presence of some particular analyte in the local environment. Understanding the relationship between this physical change and the ensuing light diffraction change is important in the design of the present invention. 35

A side-view of the array and the resulting (forward) diffraction orders is depicted in Figure 11. Lower case lambda (λ) is the wavelength of the incoming light, and upper case lambda (Λ) represents the bump center to center spacing. Incident monochromatic light with wavelength λ is diffracted into several orders with each particular order m characterized by the diffraction angle ϕ (sub m) equals $\arcsin(\lambda \times m) / \Lambda$, for m equals 0, ± 1 , ± 2 , . . . , $\pm \Lambda / \lambda$.

As can be concluded from Figure 11, no sensing information is obtained by tracking the angle of diffraction; however, changes in the bump size and shape will be reflected in the relative intensities of the diffraction orders. A change in bump spacing, however, will change the diffraction angle.

The far-field diffraction pattern for a typical micro-bump array is shown in Fig. 12. The left-hand side of Figure 12 gives a diffraction pattern for a micro-bump array with a 15 micron period, and one for a 3 micron period on the right. The brightness or irradiance of each spot is related to the statistical average physical shape, size, and refractive index of the sampled bumps. The sampled bumps include all bumps illuminated by the probe beam. The spot irradiance and the bump profile are mathematically related by the Fourier coefficients of the near field transmitted beam series expansion expression. Figure 13 graphically illustrates this relationship. The transmitted far field electric field magnitude $|T(u)|$ is periodic, because the light wave front is periodically perturbed (delayed) as it travels through the higher refractive index micro-bumps, where each order's electric field (a_0, a_1, a_2, \dots) is related to the perturbed wave front's Fourier series coefficient. Wave-fronts traveling through the bumps undergo a phase delay in proportion to the path-length traveled through the bump. The periodic phase-delay perturbation gives rise to a periodic far-field pattern. The far-field electric field magnitude of the diffraction pattern spots are related to the bump profile, $h(x)$, through the near-field transmitted electric

field, $t(x)=\exp(j kh(x))$, where $k=2\pi n/\lambda$, where n is the refractive index of the bump region. The Fourier coefficients, and hence the square root of the detected irradiance for each order are calculated by the overlap integral: $a_{(sub\ m)}=(1/\Lambda) \int_{-\Lambda/2}^{+\Lambda/2} t(x)\cos(mkx) dx$, where R is the quantity $t(x)\cos(mkx)$ integrated from $-\Lambda/2$ up to $+\Lambda/2$, where it is assumed that $t(x)$ is a symmetric function. If $t(x)$ is not symmetric, one must also calculate the overlap integral of $t(x)$ with the odd function, $\sin(mkx)$. This Fourier series coefficient expression is what allows us to predict and relate a change in the micro-bump array diffraction pattern to a change in the bump characteristics, which in turn are related to the local analyte concentration.

An example calculation depicting the diffraction order irradiance for two truncated cosinusoidal functions of different height is shown in Fig. 14. This figure illustrates a calculated half-plane, on-axis diffraction pattern irradiance for two different micro-bump arrays with refractive index = 1.5, period $\Lambda = 15$ micrometers, bump width = 10 micrometers, bump height = 1.5 micrometers (large dots), bump height = 0.9 micrometers (small dots). The abscissa is in degrees, and the ordinate is measured in power % of total transmittal of irradiance. An individual bump profile is shown in the inset. The example diffraction spectra shown in Fig. 14 illustrates a unique and measurable relationship between the bump shape and the diffraction pattern irradiance. Thus, an analyte-induced change in the bump profile would give rise to an intensity change in one or more diffraction orders. This intensity change can be measured, and in turn, related to the presence of analyte in the local environment.

The principal drawback associated with the micro-bump array diffraction sensor is the difficulty in relating and quantifying the changes in the detected diffraction irradiance signal to an input stimulus (*i.e.*, temperature, pH, etc.). Small errors introduced at any point in the transformation from detected irradiance to temperature scale are greatly multiplied

due to the non-linear transformations. Sensitivity for such a device is highly dependent on the bump geometry and initial volume. Furthermore, since the gel has a minimum trigger temperature and since it undergoes an approximate 10 times volume change over a few degrees, the dynamic range of the sensing device of the present invention is extremely limited.

One of the goals established from the onset was that the detection results should be easily interpreted without the aid of support electronics. Therefore, it is necessary to assemble the micro-bump reactors in a pattern such that the diffracted light forms an image that is easily recognizable by the eye. Furthermore, a salient feature of the diffracted image should change to indicate detection of a pre-selected analyte in the local environment.

The present invention is a unique combination of micro-lithographic and SAM fabrication techniques that have made it possible to transform the gray-scale film printout to a surface-relief pattern in photoresist, form a stamp of the pattern, and define responsive-gel-adhering, hydrophilic-patterned regions on metallized MYLAR®. The type of hologram sensor platform produced according to the present invention is unique with a whole set of unique properties that may be advantageous when compared side by side with other types of sensors. The integration of the visual display with the sensing surface according to the present invention is an important step in reducing costs associated with support electronics.

This invention is further illustrated by the following examples, which are not to be construed in any way as imposing limitations upon the scope thereof. On the contrary, it is to be clearly understood that resort may be had to various other embodiments, modifications, and equivalents thereof, which, after reading the description herein, may suggest themselves to those skilled in the art without departing from the spirit of the present invention.

Example 1

Printing of nickel/gold-coated MYLAR® (polyethylene terephthalate) with patterns of 16-mercaptohexadecanoic acid and hexadecanethiol

A nickel/gold alloy of 15.9 nm thickness was sputter-coated onto 7 mil MYLAR®. The composition had 65% visible light transmittance, and 65 ohms/cm² resistance. The following results from XPS surface analysis were obtained.

<u>Sputter Time</u> <u>(sec)</u>	<u>%C</u>	<u>%O</u>	<u>%Au</u>	<u>%Ni</u>
0	51.5	8.0	40.5	ND
15	33.3	6.4	60.3	ND
30	20.2	ND	71.7	8.0
60	19.3	ND	72.4	8.3

ND means "not detected", i.e., less than 0.2 atom-percent.

These results show that the outermost surface of the Ni/Au alloy is predominantly Au, i.e., Ni is not detected until after approximately 5.0 nm of Au is removed. Thus, the alloy presents a surface that resembles pure gold and can be used as a "pure gold" surface for contact printing.

MYLAR® film modified with a sputter-deposited nickel/alloy topcoat was obtained from Courtaulds Performance Films (21034 Osborne Street, Canoga Park, CA 91304). Patterns of hydrophilic, carboxy-terminated alkane thiols were stamped onto the Ni/Au metallized MYLAR® using CH₃(CH₂)₁₅SH and HOC(O)(CH₂)₁₅SH acid by the following method. (See Figure 1). An exposed and developed photoresist pattern of 10 micron diameter circles on a silicon wafer was used as the master. Polydimethylsiloxane (PDMS; silicone elastomer 184; Dow Corning Co., Midland, MI), was polymerized on a master to produce a stamp with ten micron-diameter circles spaced five microns apart. The stamp was

inked by exposure to a solution (1 to 10 mM in ethanol) of 16-mercaptohexadecanoic acid, and allowed to air-dry. The substrate was contacted with the stamp for 50 seconds and washed for 2 to 4 seconds with a solution of hexadecanethiol (1 to 10 mM in ethanol) which reacts with the gold in regions not covered by the carboxy terminated thiol. A patterned surface with CO₂H and CH₃ is thus produced. The substrate was finally washed for 10 seconds in ethanol and dried in a stream of nitrogen. The results of this printing are shown in Figure 2.

These hydrophilic self-assembling monolayer circles allow for selective placement of high surface tension fluids such as water, triethylene glycol, or ultraviolet light curable urethane acrylic adhesives. Aqueous solutions of responsive gels with or without indicators sensitive to analytes can also be placed selectively on the monolayer circles. These liquids can contain dissolved and suspended reagents that react chemically or physically with targeted analytes, thus making the coated plastic film a collection of 10 micron microreactors suitable for low-cost, disposable chemical sensors. An example of such a device is shown in Figure 3a.

Diffraction of visible light was shown with these compositions. Both reflected and transmitted diffraction patterns were observed when using 5mW, 670 nm laser illumination. Figure 3b is a photograph of the diffraction pattern formed by visible light shown through the self-assembling monolayer pattern of Figure 3a. Rainbow diffraction colors were observed with transmitted white light.

Measurement of Contact Angles

Contact angles were measured on a Rame-Hart Model 100 goniometer at room temperature and ambient humidity. Water for contact angles was deionized and distilled in a glass and Teflon apparatus. Advancing and receding contact angles were measured on both sides of at least three drops of each

liquid per slide; data in the figures represents the average of these measurements. The following method was used for measuring contact angles: A drop approximately 1-2 microliters in volume was grown on the end of a pipette tip (Micro-Electrapette syringe; Matrix Technologies; Lowell, MA). The tip was then lowered to the surface until the drop came in contact with the surface. The drop was advanced by slowly increasing the volume of the drop (rate approximately 1 microliter/second). Advancing contact angles of water were measured immediately after the front of the drop had smoothly moved a short distance across the surface. Receding angles were taken after the drop had smoothly retreated across the surface by decreasing the volume of the drop.

X-ray Photoelectron Spectroscopy (XPS)

X-ray photoelectron spectra were collected on a Surface Science SSX-100 spectrometer using a monochromatized Al K-alpha source ($h\nu=1486.6$ electron volts). The spectra were recorded using a spot size of 600 micrometers and a pass energy on the detector of 50 electron volts (acquisition time for one scan was approximately 1.5 minutes). For the monolayers, spectra were collected for carbon and oxygen using the 1s peaks at 285 and 530 eV, respectively; the binding energies for elements in the monolayer were referenced to the peak due to hydrocarbon in the C 1s region, for which the binding energy was fixed at 284.6 eV. Spectra for the solid hydroxamic acid were collected using an electron flood gun of 4.5 eV to dissipate charge in the sample. The binding energies for the substrates were not standardized to a reference sample. All spectra were fitted using an 80% Gaussian/20% Lorentzian peak shape and a Shirley background subtraction. See J.P. Folkers, G.M. Whitesides, *et al.*, *Langmuir*, vol. 11, no. 3, pp. 813-824 (1995).

Condensation Figures

Condensation figures (CFs) are arrays of liquid drops that form upon condensation of vapor onto high surface energy regions of a patterned solid surface. The examination of condensation figures has historically been used as a method to characterize the degree of contamination on an otherwise homogeneous surface. One is able to impose a pattern on arrays of condensed drops by patterning the surface underlying them into regions of different solid-vapor interfacial free energy and to characterize the patterned CFs by photomicroscopy and optical diffraction. It can be demonstrated that appropriately patterned CFs can be used as optical diffraction gratings and that examination of the diffraction patterns provides both a rapid, nondestructive method for characterizing patterned self-assembling monolayers and an approach to sensing the environment (see Figure 6, explained below). Because the form of the CFs—that is, the size, density, and distribution of the drops is sensitive to environmental factors, CFs of appropriate size and pattern diffract light and can be used as sensors. This principle is demonstrated by correlating the temperature of a substrate patterned into hydrophobic and hydrophilic regions, in an atmosphere of constant relative humidity, with the intensity of light diffracted from CFs on these regions.

Appropriate patterns are formed from self-assembled monolayers (self-assembling monolayers) on gold/nickel by using combinations of hexadecanethiol [$\text{CH}_3(\text{CH}_2)_{15}\text{SH}$], 16-mercaptohexadecanoic acid [$\text{HS}(\text{CH}_2)_{15}\text{COOH}$], and 11-mercaptopundecanol [$\text{HS}(\text{CH}_2)_{11}\text{OH}$]. Several techniques are now available for preparing patterns of two or more self-assembling monolayers having 0.1- to 10- μm dimensions.

At 20°C, an incident beam of light from a laser (helium-neon laser, wavelength = 632.8 nm) produced a single transmitted spot because no water had condensed on the surface, and the transmittance of the regions covered with

different self-assembling monolayers were effectively indistinguishable. As the surface was exposed to warm, moist air, droplets of water condensed preferentially on the hydrophilic regions. Diffraction patterns appeared in the light transmitted from the surface. Under these conditions, light was transmitted coherently from the regions where no water had condensed and was scattered by the regions where water had condensed. The condensation figures disappeared within several seconds as the water droplets which condensed on the self-assembling monolayers evaporated.

The ability to form condensation figures can be ascertained by the relative contact angles of water on the hydrophobic and hydrophilic self-assembling monolayers. Unpatterned monolayers of the appropriate thiol were prepared by immersion of the substrate in a dilute solution for one hour, followed by rinsing with ethanol and air drying.

The contact angles of water on Au(Ni)/MYLAR® reacted with $\text{CH}_3(\text{CH}_2)_{15}\text{SH}$ and $\text{HOC}(\text{O})(\text{CH}_2)_{15}\text{SH}$ were 100° and 62° , respectively. The untreated Au(Ni)/MYLAR® contact angle for water was $73-77^\circ$. This water contact angle is similar to that obtained for Au coated SiO_x wafers, which is $73-74^\circ$ (data not shown).

Condensation Figures [*Science*, Vol. 263, 60 (1994), incorporated herein by reference] with equivalent optical diffraction can be formed on Au/Ni:MYLAR®, relative to known art with Au:SiO_x. The chemistry of alkane-thiols reacting with Au/Ni:MYLAR® is similar to that reported in the literature for Au:SiO_x.

A field emission secondary electron microscope image of 10 micron-diameter circles of hydrophilic self-assembling monolayers formed by printing of 16-mercaptohexadecanoic acid onto MYLAR® metallized with Ni/Au alloy is shown in Figure 2. Figure 3a is an optical photomicrograph at 300x magnification of 10 micron-diameter circles of hydrophilic self-assembling monolayers formed by printing of 16-

mercaptohexadecanoic acid, and after exposure to a high surface energy, curable, optical adhesive. The adhesive was cured by ultraviolet light (UV) exposure.

5 Figure 3b is a photograph of the diffraction pattern formed by visible light shown through the self-assembling monolayer pattern described by Figure 3a.

10 Figure 4 is a field emission secondary electron micrograph images of 10 micron-diameter circles formed of self-assembled photocurable polymers on hydrophilic self-assembling monolayers.

15 Figure 6, in the left hand vertical column, shows printed arrays, on the right-hand column shows the resulting diffraction patterns. The inset in the left hand column in Figure 6 D gives the scale of the patterns.

Example 2

Placing a hydrogel atop patterns of self-assembling monolayers printed on nickel/gold-coated MYLAR® (polyethylene terephthalate)

20 Solutions of a chemically responsive gel were prepared by polymerizing an aqueous solution of N-isopropylacrylamide (NIPA) (18 weight %) using persulfate and ascorbic acid. Briefly, a 250 mL Erlenmeyer flask was charged with 18.0 gm NIPA monomer and 80g distilled water. The resulting
25 solution was stirred using a magnetic stir bar, and 0.08 g potassium persulfate was allowed to dissolve. A rubber septa was placed on the flask, and inlet and outlet needles were used to purge the flask with nitrogen. Stirring constantly, the flask was cooled in an ice bath to approximately 0°C. A solution of
30 0.07 g L-ascorbic acid in 3 mL distilled water was injected into the mixture. Within 2 hours, the mixture had polymerized to a very viscous, clear solution. The resulting polymer was diluted with additional water to a concentration of 2 percent by weight. To this solution was added an equal
35 volume of toluene to produce a two phase system. The printed

MYLAR® film of **Example 1** was drawn through both phases of this system to self-assemble the aqueous solution of polymer onto the patterned, hydrophilic SAM. This resulted in a sensing device containing a stimuli responsive gel.

Example 3

Printing of gold-coated MYLAR® (polyethylene terephthalate) with patterns of 16-mercaptohexadecanoic acid and hexadecanethiol

Patterns of gold-coated MYLAR® (polyethylene terephthalate) were printed with patterns of 16 mercaptohexadecanoic acid and hexadecanethiol, in a manner similar to that shown in Figure 1, and described below.

MYLAR® film modified with a plasma deposited gold topcoat was obtained from Courtaulds Performance Films (Canoga Park, CA 91304). An atomic force microscopy image of this MYLAR® film is shown in Figure 5. Polymer film thickness between 2 and 7 mils and gold topcoats producing a surface resistance of 65 ohms per square centimeter with a visible light transmittance between 20% and 65% were used.

Patterns of hydrophilic, carboxy-terminated alkane thiols were stamped onto gold-coated film using 16-mercaptohexadecanoic acid by the following method. An exposed and developed photoresist pattern of 10 micron diameter circles on a silicon wafer was used as the master. Polydimethylsiloxane (PDMS; silicone elastomer 184; Dow Corning Co., Midland, MI), was polymerized on a master to produce a stamp with ten micron-diameter circles spaced five microns apart. The stamp was inked by exposure to a solution (1 to 10 mM in ethanol) of 16-mercaptohexadecanoic acid, and allowed to air-dry. The substrate was contacted with the stamp for 50 seconds and washed for 2 to 4 seconds with a solution of hexadecanethiol (1 to 10 mM in ethanol). The substrate was

finally washed for 10 seconds in ethanol and dried in a stream of nitrogen. (Results not shown).

Example 4

5 *Experimental Diffraction-measurement*

 An experimental diffraction-measurement schematic is shown in Fig. 15. This figure shows an experimental setup for quantifying the diffraction spectra from a diffraction array. The elements of the set-up include a Helium/Neon
10 laser, a remote shutter, a polarizer, a neutral density filter, a mirror, and expander, an iris, a sample, a lens, a Fourier transformed lens, a zoom lens, a CCD camera, a monitor, a frame grabber, and a computer with image analysis software.

 Coherent light with wavelength $\lambda = 0.6328$ micrometers
15 was diffracted by the sample which was enclosed in an environment chamber. The diffraction orders were collected by a lens and imaged onto a CCD camera where the image was displayed on a monitor and stored on a computer for image processing and analysis. This experimental setup was used to
20 measure the two dimensional diffraction image intensity from a self-assembled-monolayer (SAM)-patterned array of temperature-responsive poly-N-isopropylacrylamide (NIPA). The poly-NIPA bumps undergo a volume shrinkage with increasing temperature, therefore, the detected diffraction
25 order irradiance from one spot to the next changed, as was predicted by the model, with a rise in temperature. An example image captured during this experiment is shown below in Fig. 16. The zero order beam in this figure was blocked to eliminate CCD saturation. S. Hirotsu, Y. Hirokawa,
30 and T. Tanaka, "Volume-phase transitions of ionized N-isopropylacrylamide gels," J. Chem. Phys., vol. 87, no. 2, pp. 1392-1394, July 15, 1987, incorporated herein by reference.

 Conversion from the diffraction pattern's gray-scale
35 image to temperature scale must be done with extreme care

because it involves at least five sequential transformations including: (1) a non-linear relationship between the gel temperature and the micro-bump volume, (2) a relationship between the bump volume and shape, (3) a non-linear (oscillating) relationship between the bump shape and the signal detected with the CCD device (see Fig. 17) and finally, (4) scaling and quantization of the detected signal for digital storage of the image.

Assuming the bump is cosinusoidal shaped with height h , and constant base diameter d , the volume v , of the microbump is $v = hd^2(1 - 2/\pi)$, i.e., the volume is linearly related to the height for this shape. Figure 17 is a graph of the first-order diffraction irradiance as a function of the micro-bump height for a bump refractive index of 1.5, base width of 10 micrometers, and a period of 15 micrometers. The oscillating irradiance curve is linear in a finite range, and limits the useful bump height to approximately 0.3 micrometers for this geometry. The abscissa measure bump height in microns, and the ordinate measures irradiance in power % of total transmittal of irradiance.

Example 5

Holographic pattern design and method

A novel hologram synthesis method compatible with contact printing and SAM (see A. Kumar, G. Whitesides, *et al.*, cited *supra*) technology was developed. The hologram computation is based on traditional holography theory with adjustments made to account for physical considerations such as printer resolution, wavelength scaling, sampling theory, and image content. The recent development and availability of high-resolution film printers is one of key enabling technologies which has allowed rapid refinement cycles and cost effective hologram synthesis. It should be stressed that the diffracted image quality is directly limited by the printer resolution, i.e., by how small and close together individual

dots can be placed in the printer output. A 3600 dot per inch specification means that the minimum resolution the printer is capable of producing is 7 micrometer diameter dots with a 7 micrometer center- to center spacing. To put this into perspective, standard holographic silver halide films range in grain size from 0.05 micrometers to 1.0 micrometers. The table below summarizes the "rule of thumb" resolution requirements for embossed holograms of various quality. One must be aware that the 7 micrometer feature-size is huge in terms of traditional display holography and severe limitations are imposed by such resolution, nevertheless, a limited class of special thin transmission holograms can be readily made with such resolution, enabling low-cost development and testing of the holographic diffraction sensor.

"Rule of thumb" resolution requirements for embossed holograms of various quality

EMBOSSSED IMAGE QUALITY	DENSITY (pixels/mm)	FEATURE SIZE (μ meters)
Very High Quality 3D images	14,000	< 0.7
Medium Quality 3D images	~ 1,000	~ 1.0
Good 2D, 3D images	~ 600	~ 1.7
Good 2D, Poor 3D images	< 450	> 2.0

Figure 18 depicts the geometry for a traditional holographic transparency recording setup. The figure shows an object plane, a film plane, a signal beam, and a reference beam, with a smiling face as the holographic image. Cartesian coordinates are also shown, with (0,0,0) as the reference point, and Φ as the angle the reference beam makes with the z axis in the x-z plane. The object plane consists of small transparent holes in an opaque background. The signal and reference beams are assumed mutually coherent and monochromatic. The light propagating from each point in the object plane can be expressed, using Huygens principle, as an expanding

spherical wave with an electric field phasor representation:
 $E(r) = \exp(jkr)/r$, where $k = 2\pi/\lambda$ and $j = \sqrt{-1}$. The reference
 plane-wave beam is incident on the film plane at an angle $\phi_{\text{sub(ref)}}$
 with respect to the film normal. The image beam and
 the reference beam interfere and the constructive/destructive
 interference pattern is recorded in the film. Setting $\phi_{\text{sub(ref)}} = 0$
 reduces the resolution requirements of the film but
 an on-axis hologram is produced. The drawbacks of the
 on-axis hologram will be discussed subsequently.

The holographic recording setup in Fig. 18 can be
 modeled on a computer and an interference pattern at the film
 plane can be calculated, converted to a gray-scale bit map
 image, and printed out on a high-resolution film printer. The
 result is a computer generated transparency hologram. Each of
 the N object points (*i.e.*, the dots making up the smile-face
 image in Fig. 18) are expressed in Cartesian coordinates as
 (x_0, y_0, z_0) . Each point can be assigned a unique position,
 therefore, 3-dimensional object representation is allowed. Each
 point in the film plane is expressed as (x_f, y_f, z_f) where z_f
 is the minimum distance from the object to the film plane, and it
 is usually a fixed value, thereby denoting a flat piece of
 recording film. A convenient zero-phase reference point in
 the film plane is picked and designated (x_{f0}, y_{f0}, z_{f0}) . The
 minimum distance from each point on the object to an
 arbitrary point on the film is:

$$r = [(x_0 - x_f)^2 + (y_0 - y_f)^2 + (z_0 - z_f)^2]^{1/2}$$

The radial distance to the zero- phase reference point is:

$$r_0 = [(x_0 - x_{f0})^2 + (y_0 - y_{f0})^2 + (z_0 - z_{f0})^2]^{1/2}$$

The difference between the r and r_0 , scaled by the propagation
 constant, k , gives the differential phase of the image wave for

each point on the film plane. The main hologram synthesis equation is:

$$H(x_f, y_f) = \sum \exp[jk(r-r_0) - x \sin(\phi_{\text{sub}}(\text{ref}))]$$

5

summed from $n=1$ to N . The synthesis equation is used in calculating the contribution of each N points comprising the object and it is solved at each sample point (x_f, y_f, z_f) in the film plane. If the film plane is to be represented by an $M \times M$ array of sample points, then the number of synthesis calculations required is $N \times M^2$.

10

The synthesized hologram consists of a two-dimensional array of sample points representing the complex phase and amplitude of the interference pattern between object and reference beams. Conversion of the complex array values to a gray-scale must be done before printing the hologram pattern because only the amplitude information can be represented by the printer.

15

Since the imaginary part of the information is discarded in the conversion, it makes no sense to calculate it in the first place. Therefore, a more efficient and direct synthesis equation is:

20

$$H(x_f, y_f) = \sum \cos[jk((r-r_0) - x \sin(\phi_{\text{sub}}(\text{ref})))]$$

25

where the summation is from $n=1$ to N . This synthesis equation can be implemented with a computer, for example, the MathCad program (MathSoft, Cambridge MA).

30

Example 6

Sampling Requirements

35

Care must be taken to calculate enough points on the hologram interference pattern (*i.e.*, to sample at a sufficiently high spatial frequency) so that the information is preserved and aliasing does not occur. A form of the Shannon or Nyquist

sampling theorem can be applied to the case at hand. A. V. Oppenheim and R. W. Schaffer, Discrete time signal processing (Prentice-Hall, Englewood Cliffs, N.J. 1989), incorporated by reference. However, a more direct and physically appealing method was devised to gain insight into the problem and to assist with the design.

Since the calculated hologram consists of periodic sample points which, upon printout, are separated by a minimum center-to-center spacing of $\Lambda = 7$ micrometers (due to the resolution of the printer), the reconstructed image will also be repeated periodically in space with an angular repetition (recall Fig. 11) equal to:

$$\theta_m = \sin^{-1}(\lambda x / \Lambda)$$

which is approximately 5 degrees for red light incidence ($\lambda = 0.6328$ micrometers). This means that the full image fan angle can not exceed 5 degrees without overlapping with the replicated images. Full use of the image plane is possible only when the printer resolution is equal to the incident wavelength, and when the all the complex phase information is retained. The limited printer resolution ultimately limits the extent of the image and specifies the minimum image projection distance from the film for a given image extent.

Example 7

Conjugate Beam Aberrations

Another consideration that plays an important role in the hologram design is the unwanted twin or conjugate beam that arises due to the inability to print all the phase information. There are two possible objects giving rise to the same hologram pattern due to the uncertainty of π in recording phase: one is the original object, the other is a virtual object located symmetrically on the other side of the film plane. This concept is illustrated in Fig. 19. Figure 19 illustrates the twin

images arising from the hologram, the virtual image on the left-hand side of Figure 19, and the real image on the right-hand side, with the viewer positioned at the lower right-hand corner.

5 A hologram of a single point consists of symmetric rings on the film plane spaced finer and finer with increased distance from the point center. Figure 11 is a side-view of the bottom half-plane hologram pattern and it portrays the incident beam, the transmitted beam, and two diffraction orders for each
10 incident ray. The angle of diffraction is set by the local period, just as illustrated in Fig. 11. One set of diffraction orders comprise the real image and converge to a point to the right of the film plane. The other set of orders diverge from the film plane in a pattern that appear to the eye to be coming from a
15 point located at the virtual image. Figure 20 illustrates conjugate beam haze. The in-focus image is the real image, the large dot is the transmitted beam, and the haze is the virtual image. The projected image on the left is illuminated through hologram center. Image on right is illuminated slightly
20 off-center to illustrate the symmetric conjugate image placement. This figure demonstrates the effect of the twin beam aberration for an example real image. In viewing the image of the real object, one has to look through an out-of-focus background image of the virtual object, a most
25 annoying disturbance, and one of the problems that plagued the first holograms made by Dennis Gabor in 1948. D. Gabor, "A new microscopic principle," Nature, vol. 161, pp. 777-778 (1948).

30 In the early 1960's, Leith and Upatnieksi discovered that the twin-beam problem could be alleviated by modulating the signal beam on a carrier to spatially separate the real and virtual image beams. E. N. Leith and J. Upatnieks. "Wavefront reconstruction with diffused illumination and three-dimensional objects." J. Opt. Soc. Am, vol. 53, pp. 1377-1381
35 (1964), incorporated herein by reference. This so-called

"off-axis hologram" geometry is accomplished either by centering the object and shifting the reference beam angle, or equivalently, by setting the reference beam to zero and shifting the object off axis. Care must be taken to insure that the combined reference beam angle and the image cone angle are within the limited angular range defined by the sampling period.

Example 8

Fabrication

The challenge one faces in the fabrication process is to faithfully reproduce the hologram features and scale at a reasonable cost. In this Example, an overview is presented of the fabrication steps. The ability to "print" a hologram with responsive material is an important factor in the fabricating a sensing device according to the present invention. The use of MYLAR® as a hologram base substrate is important to high volume, continuous processing, and cost per unit reduction.

The present invention has been developed to enable the positioning of responsive-material in the form of a hologram pattern onto metallized MYLAR®. The steps include: (1) printout of the computer bit map hologram pattern to high-resolution transparency film, (2) photolithographic transfer of the mask pattern to photoresist, (2) formation of an elastomeric stamp from the photoresist relief pattern, (3) thiol monolayer patterning on MYLAR® using the stamp, and (4) responsive gel assembly on the pattern defined by the thiol monolayer.

Example 9

Computer bitmap

The hologram generated by the computer is initially stored in the form of a 256-level gray-scale bitmap pattern. Each pixel in the bitmap, therefore, is represented by one byte of information. The image storage requirements are set by the total number of samples. For example, a 1000 x 1000 dot gray-scale bitmap occupies approximately 1 megabyte of

memory. An example of the type of hologram pattern that is produced from a recognizable object is shown below in Fig. 21. The recognizable object is on the left-hand side, and its associated hologram is on the right-hand side. The figure on the right is what is patterned. It is a gray-scale representation of the pattern that is produced from the hologram synthesis equation using the image points shown in the figure on the left.

Once the hologram bitmap is calculated, it is converted to a tagged image format (.tif) file and imported into a graphics application such as CorelDraw (Ottawa, Ontario Canada), which is capable of converting the image file to a printer file. Nearly all of the prototype holograms produced for our project were formatted for a 3600 dpi resolution Agfa Selectset 5000 film printer (Ridgefield Park, NJ).

Several different synthesized holograms can be placed on the same page to minimize the cost per test. Figure 22 shows a 3600 dots per inch (dpi) test page printout with 20 test holograms on one page. A typical printer file for a multi-test run contains approximately 100 megabytes of information: therefore, a removable "Zip" drive may be utilized to conveniently transfer the file to the service bureau for printout.

The hologram mask is inspected for image quality in both transmission and projection mode before attempting to transfer the pattern to the photoresist-covered substrate. The ability to view the holographic image on the printer-output film saves time in the development process. Images can be evaluated for sampling density, extent limits, and depth before the transfer process is carried out.

Example 10*Photoresist preparation and patterning*

5 The next step in the process is to coat a suitable flat, polished substrate with 1-2 micrometers of photoresist, expose to ultraviolet (UV) light, and develop according to the following procedure):

1. Clean substrate with methanol.
2. Rinse with de-ionized (DI) water.
- 10 3. Clean with acetone.
4. Rinse with DI water.
5. Clean with trichloroethane (TCE).
6. Rinse with DI water.
7. Blow off excess with nitrogen stream.
- 15 8. Spin to remove any excess water.
9. Bake at 100° C for 15 minutes to remove water.
10. Pool hexamethyldisilane on wafer and spin at 5000 revolutions per minute (rpm) for 25 seconds.
11. Pool photoresist (Shipley SC1857) on wafer and spin at 5000 for 25 seconds.
- 20 12. Bake at 120° C for 20 minutes. (Don't over bake or photoresist will become brittle).
13. Expose photoresist through mask at approximately 85 mJ/cm² at 338 nanometers wavelength.
- 25 14. Develop with 1:1 (DI water: Shipley concentrate) for 10 seconds to remove exposed photoresist.

30 The resulting relief-patterned substrate is denoted the "master" and it is the mold from which the elastomeric stamp is formed. An additional step of "de-scumming" the relief wells with a reactive ion etch may be necessary if the bottom of the wells contain any residual photoresist.

Example 11*Elastomeric stamp formation*

After the master is fabricated, cleaned, and inspected, it is placed in a vacuum chamber with a small amount of fluorine compound to passivate the surface. Then an elastomer is poured on the master mold and allowed to cure for 16 hours at about 65° C. It is important to coat the surface with the passivation layer first so that the elastomer stamp can peel away from the master when it is set.

Example 12*Final hologram formation*

The elastomeric stamp is coated with a hydrophilic thiol and the hologram pattern is printed on a metallized MYLAR® sheet (via the thiol) by carefully pressing the stamp against the MYLAR® and by applying uniform pressure. An analyte-specific responsive gel can then be assembled in the regions defined by the thiol pattern by hydrophilic attraction between the gel and the thiol.

Example 13*Process examples*

There are four transfer steps involved in the hologram production. Each step can be evaluated visually for good pattern replication by microscopy. Figure 23 A-D shows several pattern transfer from the printer output on high-resolution printer film (A), to the photoresist master on gold film (B), to the elastomer stamp (C), to the responsive gel on metallized Mylar® (D). The small line in (C) is 30 microns long. These figures are corner views of the same pattern at each step. In the figures 23 A-D, the pattern transfer from the printer to the stamp would be judged to be "good" but the quality of the final transfer to the MYLAR® is difficult to judge due to the contrast of the pattern and because of the residual drops of gel which tend to obscure the image.

A similar comparison of the hologram's central region, as shown in Fig. 9, indicates a good transfer of the printed pattern to the gel on MYLAR®. The intermediate steps are not shown in this figure.

5

Example 14

Image Reconstruction

The holographic image can be viewed in one of two ways depending on the lighting source. The virtual image can be seen behind the film plane when the hologram is placed between the eye and a white light point source. Alternatively, the real image can be projected on a screen, as shown in Figure 24, by shining a laser through the hologram. The reconstructed image perspective is determined by the region of beam incidence in the film plane. The fact that each spot in the film contains a separate perspective view of the entire image is one of the most useful characteristics of the hologram, part of the film may become damaged but the image may be viewed simply by moving to a different location on the film.

Another useful attribute associated with the real image reconstruction is shift invariance. Incident light can be scanned across the film plane with no apparent shift in the image. This characteristic is very useful in fixed-position detector systems where precise positioning of the responsive hologram film is impractical.

Figure 7, at the left hand side, is an illustration of a printed hologram, and the corresponding smiling face image it produces. The right hand side of Figure 7 are two enlarged views, side plan and top plan, of the circles of self-assembling monolayers which all together make up the printed hologram.

30

Example 15

Hologram synthesis algorithm using the MathCad program

5 The MathCad Program synthesizes the interference pattern required for reconstruction of a 3 dimensional cube made of finite point sources and located behind the film plane. The dimension z is defined in the synthesis equation for each point so that depth can be given to the image.

10 $TOL = 10^{-9} \text{ mm} = 1 \times 10^{-3} \text{ micrometers} = 1 \times 10^{-6} \text{ cm}$
 $= 1 \times 10^{-2} k = 2\pi/\lambda$

$\lambda = 0.6328 \text{ micrometers}$

$F_{min} = 7.056 \text{ micrometers}$ Minimum printable feature size, i.e. minimum sample period.

15 $NS = 1000$ Number of samples in one dimension making up the hologram

$EF = F_{min}NS$

$EF = 7.056 \text{ mm}$ Maximum extent of hologram window.

20 $Z = 6 \text{ cm}$ Real image projection (in focus) distance from the film

$EI = Z \tan[\arcsin(\lambda / F_{min})]$

25 $EI = 5.403 \text{ mm}$ Extent of the projected real image without aliasing

$x_{off} = 0$ Offset of the real image from center requires a modulation.

$\Theta(R) = \arctan(x_{off}/Z)$ Required reference beam angle (from film plane normal)

30

$E_f = 7.056 \text{ mm}$

$E_i = 5.403 \text{ mm}$

$z = 60 \text{ mm}$

$\Theta(\text{ref}) = \arctan(\sigma \tan(\Theta(R)))$

35 $N = 44$ Number of points in the image

Object Point Definitions Defining a Cube

$$\mathbf{x}_0 = (0 \text{ } 2.5 \text{ } 5 \text{ } 7.5 \text{ } 10 \text{ } 10 \text{ } 10 \text{ } 10 \text{ } 10 \text{ } 7.5 \text{ } 5 \text{ } 2.5 \text{ } 0 \text{ } 0 \text{ } 0 \text{ } 0 \text{ } 0 \text{ } 0 \text{ } 0 \text{ } 0 \\ 0 \text{ } 0 \text{ } 0 \text{ } 0 \text{ } 0 \text{ } 10 \text{ } 10 \text{ } 10 \text{ } 10 \text{ } 10 \text{ } 10)$$

5 $y_0 = (0 \ 0 \ 0 \ 0 \ 0 \ 2.5 \ 5 \ 7.5 \ 10 \ 10 \ 10 \ 10 \ 10 \ 7.5 \ 5 \ 25 \ 0 \ 0 \ 0 \ 0 \ 2.5$
 $5 \ 7.5 \ 10 \ 10 \ 10 \ 10 \ 0 \ 0 \ 0 \ 0 \ 2.5 \ 5 \ 7.5)$

$$\mathbf{z}_0 = (0 \ 0 \ 0 \ 0 \ 0 \ 0 \ 0 \ 0 \ 0 \ 0 \ 0 \ 0 \ 0 \ 0 \ 2.5 \ 5 \ 7.5 \ 10 \ 10 \ 10 \ 10 \ 10 \ 7.5 \ 5 \ 2.5 \ 2.5 \ 5 \ 7.5 \ 10 \ 10 \ 10 \ 10 \ 1)$$
$$10 \quad x_0 = x_0^T(E_i/20) \quad y_0 = y_0^T(E_i/20) \quad z_0 = z_0^T(E_i/20) + z$$

Film point definitions

$$\Delta x = E_f / NS \quad \Delta y = \Delta x \quad \Delta x = 7.056 \text{ micrometers}$$

15 sampling period

$$\begin{aligned} i=0..NS-1 \quad j=0..NS-1 \quad x_i=i\Delta x - E_f/2 \\ y_j=j\Delta y - E_f/2 \end{aligned}$$

Interference pattern calculation

$$H_{i,j} = \sum \cos \{ k [[(x_i - x_{0n})^2 + (y_j - y_{0n})^2 + (z - z_{0n})^2]^{1/2} - [(x_{0n})^2 + (y_{0n})^2 + (z_{0n})^2]^{1/2} - (x_i) \sin(\theta_{ref})] \}$$

Those skilled in the art will now see that certain modifications can be made to the invention herein disclosed with respect to the illustrated embodiments, without departing from the spirit of the instant invention. And while the invention has been described above with respect to the preferred embodiments, it will be understood that the invention is adapted to numerous rearrangements, modifications, and alterations, all such arrangements, modifications, and alterations are intended to be within the scope of the appended claims. All publications cited herein are incorporated herein by reference.

Claims

What is claimed is:

- 5 1. A sensing device comprising:
 - (a) a self-assembling monolayer printed on a substrate; and
 - (b) a gel associated with the self-assembling monolayer, the gel being capable of responding to a stimulus.
- 10 2. The device of claim 1 wherein the sensing device can form a hologram when electromagnetic radiation is transmitted through the device.
- 15 3. The device of claim 1 wherein the substrate is selected from the group consisting of glass, silicon dioxide, aluminum oxide, and metallized polymer films.
- 20 4. The device of claim 1 wherein the gel is formed from N-isopropylacrylamide, or a derivative thereof.

5. The device of claim 1, wherein the gel is agarose, dextran, carageenan, alginic acid, starch, cellulose, deionized gelatin, polyvinyl alcohol, polyacrylic acid, polyacrylamide, or polyethylene glycol, N-vinyl pyrrolidone, glycidyl methacrylate, hydroxyalkyl acrylates, hydroxyalkyl methacrylates, hydroxyethyl acrylate, hydroxypropyl acrylate, hydroxybutyl methacrylate; epoxy acrylates, epoxy methacrylates, glycidyl methacrylate; amino alkyl acrylates, amino alkyl methacrylates, N-vinyl pyrrolidone, N-vinyl carbazole, N-vinyl acetamide, N-vinyl succinimide, amino styrenes, polyvinyl alcohols, polyvinyl amines, N-isopropyl acrylamide, vinyl pyridine; vinyl sulfonate polyvinyl sulfate; vinylene carbonate; vinyl acetic acid, vinyl crotonic acid; allyl amine, allyl alcohol, or vinyl glycidyl ethers.

6. The device of claim 1, wherein the gel is an organogel.

7. The device of claim 6, wherein the organogel is aluminum stearate, oleate, naphthenate, or electroconductive gels, such as alkyl thiophenes.

8. The device of claim 1, wherein the gel is derivatized to contain hydroxyl, carboxyl, amino, aldehyde, carbonyl, epoxy, crown, or vinyl groups.

9. The device of claim 1, wherein the gel contains chromophores, metal salts, ions, antibodies, T or B cell receptors, fragments, or epitopes thereof, proteins, peptides, neurotransmitters, hormones, growth factors, cytokines, monokines, lymphokines, nutrients, enzymes, receptors, macromolecular structures, organelles, cells, or microorganisms.

10. The device of claim 1, wherein the gel contains compounds selected from the group consisting essentially of metallo-phthalocyanines, surfactants, NaBr, KBr, NaCl, KCl, NaI, and KI, methanol and glycerol, tetra-alkylammonium bromides, crown ethers, benzo[18]crown-6, and azobenzene chromophores.

11. The device of Claim 1, wherein there are two or more self-assembling monolayers with different physical or chemical properties.

12. The device of Claim 1, wherein a first self-assembling monolayer is hydrophobic, and a second self-assembling monolayer is hydrophilic.

13. The device of Claim 1, wherein the self-assembling monolayer is formed from compounds with the following general formula:

X-R-Y

wherein:

X is reactive with the metal or metal oxide on the polymer film;

R is a hydrocarbon chain; and

Y is a compound with any property of interest.

14. The device of Claim 13, wherein:

X is a asymmetrical or symmetrical disulfide (-R'SSR, -RSSR), sulfide (-R'SR, -RSR), diselenide (-R'Se-SeR), selenide (R'SeR, -RSeR), thiol (-SH), nitrile (-CN),
5 isonitrile, nitro (-NO₂), selenol (-SeH), trivalent phosphorous compounds, isothiocyanate, xanthate, thiocarbamate, phosphine, thioacid or dithioacid, carboxylic acids, hydroxylic acids, and hydroxamic acids;

10 R and R' are hydrocarbon chains which may optionally be interrupted by hetero atoms, and which may optionally be perfluorinated, and which are preferably non-branched; and

15 Y is optionally hydroxy, carboxyl, amino, aldehyde, hydrazide, carbonyl, epoxy, or vinyl groups.

15. The device of Claim 13, wherein R is greater than 7 carbon atoms in length.

20 16. The device of Claim 13, wherein R is a compound of the form (CH₂)_a-Z-(CH₂)_b, wherein a≥0, b≥7, and Z is any chemical functionality of interest.

25 17. The device of Claim 16, wherein Z is selected from the group consisting of sulfones, lactams, and urea.

18. The device of claim 3 wherein the substrate is a metallized polymer film, the polymer film comprising polyethylene-terephthalate, acrylonitrile-butadiene-styrene, acrylonitrile-methyl acrylate copolymer, cellophane, 5 cellulose polymers such as ethyl cellulose, cellulose acetate, cellulose acetate butyrate, cellulose propionate, cellulose triacetate, cellulose triacetate, polyethylene, polyethylene - vinyl acetate copolymers, ionomers (ethylene polymers) polyethylene-nylon copolymers, polypropylene, methyl 10 pentene polymers, polyvinyl fluoride, or aromatic polysulfones.

19. The device of Claim 3, wherein the metallized polymer film is optically transparent.

20. The device of Claim 3, wherein the metallized polymer film is metallized with metals comprising gold, silver, nickel, platinum, aluminum, iron, copper, zirconium, or alloys thereof.

21. The device of Claim 1, wherein the stimulus comprises temperature, solvent composition, mechanical strain, electric field, pH, salt concentration, pH, solvent quality, light intensity, light wavelength, pressure, ionic strength, ion identity, or specific chemical triggers.

22. The device of claim 1, wherein the hologram changes to a second hologram upon exposure to the stimulus.

23. A method of making a sensing device comprising stamping a pattern of self-assembling monolayers onto a substrate, and coating the pattern of self-assembling monolayers with one or more gels, the gels 35 being capable of responding to a stimulus.

24. The method of claim 23, wherein the device can form a hologram when electromagnetic radiation is transmitted through the method.

5

25. The method of claim 23, wherein the substrate is selected from the group consisting of glass, silicon dioxide, aluminum oxide, and metallized polymer films.

10

26. The method of claim 23, wherein the gel is formed from N-isopropylacrylamide, or a derivative thereof.

15

27. The method of claim 23, wherein the gel is agarose, dextran, carageenan, alginic acid, starch, cellulose, deionized gelatin, polyvinyl alcohol, polyacrylic acid, polyacrylamide, or polyethylene glycol, N-vinyl pyrrolidone, glycidyl methacrylate, hydroxyalkyl acrylates, hydroxyalkyl methacrylates, hydroxyethyl acrylate, hydroxypropyl acrylate, hydroxybutyl methacrylate; epoxy acrylates, epoxy methacrylates, glycidyl methacrylate; amino alkyl acrylates, amino alkyl methacrylates, N-vinyl pyrrolidone, N-vinyl carbazole, N-vinyl acetamide, N-vinyl succinimide, amino styrenes, polyvinyl alcohols, polyvinyl amines, N-isopropyl acrylamide, vinyl pyridine; vinyl sulfonate polyvinyl sulfate; vinylene carbonate; vinyl acetic acid, vinyl crotonic acid; allyl amine, allyl alcohol, or vinyl glycidyl ethers.

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28. The method of claim 23, wherein the gel is an organogel.

35

29. The method of claim 28, wherein the organogel is aluminum stearate, oleate, naphthenate, or electroconductive gels, such as alkyl thiophenes.

30. The method of claim 23, wherein the gel is derivatized to contain hydroxyl, carboxyl, amino, aldehyde, carbonyl, epoxy, crown, or vinyl groups.

5

31. The method of claim 30, wherein the gel contains chromophores, metal salts, ions, antibodies, T or B cell receptors, fragments, or epitopes thereof, proteins, peptides, neurotransmitters, hormones, growth factors, cytokines, monokines, lymphokines, nutrients, enzymes, receptors, macromolecular structures, organelles, cells, or microorganisms.

10

32. The method of claim 23, wherein the gel contains compounds selected from the group consisting essentially of metallo-phthalocyanines, surfactants, NaBr, KBr, NaCl, KCl, NaI, and KI, methanol and glycerol, tetra-alkylammonium bromides, crown ethers, benzo[18]crown-6, azobenzene chromophores.

15

20

33. The method of Claim 23, wherein there are two or more self-assembling monolayers with different physical or chemical properties.

25

34. The method of Claim 23, wherein a first self-assembling monolayer is hydrophobic, and a second self-assembling monolayer is hydrophilic.

35. The method of Claim 23, wherein the self-assembling monolayer is formed from compounds with the following general formula:

5 X-R-Y

wherein:

X is reactive with the metal or metal oxide on the polymer film;

10 R is a hydrocarbon chain; and

Y is a compound with any property of interest.

36. The method of Claim 35, wherein:

15 X is a asymmetrical or symmetrical disulfide (-R'SSR, -RSSR), sulfide (-R'SR, -RSR), diselenide (-R'Se-SeR), selenide (R'SeR, -RSeR), thiol (-SH), nitrile (-CN), isonitrile, nitro (-NO₂), selenol (-SeH), trivalent phosphorous compounds, isothiocyanate, xanthate, thiocarbamate, phosphine, thioacid
20 or dithioacid, carboxylic acids, hydroxylic acids, and hydroxamic acids;

R and R' are hydrocarbon chains which may optionally be interrupted by hetero atoms, and which may optionally be perfluorinated, and which are preferably non-
25 branched; and

Y is optionally hydroxy, carboxyl, amino, aldehyde, hydrazide, carbonyl, epoxy, or vinyl groups.

37. The method of Claim 35, wherein R is greater
30 than 7 carbon atoms in length.

38. The method of Claim 35, wherein R is a compound of the form (CH₂)_a-Z-(CH₂)_b, wherein a≥0, b≥7, and Z is any chemical functionality of interest.

35

39. The method of Claim 38, wherein Z is selected from the group consisting of sulfones, lactams, and urea.

5 40. The method of claim 23 wherein the substrate is a metallized polymer film, the polymer film comprising polyethylene-terephthalate, acrylonitrile-butadiene-styrene, acrylonitrile-methyl acrylate copolymer, cellophane, cellulosic polymers such as ethyl cellulose, cellulose acetate, cellulose acetate butyrate, cellulose propionate, cellulose triacetate, cellulose triacetate, polyethylene, polyethylene -
10 vinyl acetate copolymers, ionomers (ethylene polymers) polyethylene-nylon copolymers, polypropylene, methyl pentene polymers, polyvinyl fluoride, or aromatic polysulfones.

15 41. The method of Claim 23, wherein the metallized polymer film is optically transparent.

20 42. The method of Claim 23, wherein the metallized polymer film is metallized with metals comprising gold, silver, nickel, platinum, aluminum, iron, copper, zirconium, or alloys thereof.

25 43. The method of Claim 23, wherein the stimulus comprises temperature, solvent composition, mechanical strain, electric field, pH, salt concentration, pH, solvent quality, light intensity, light wavelength, pressure, ionic strength, ion identity, or specific chemical triggers.

30 44. The method of claim 23, wherein, wherein the hologram changes to a second hologram upon exposure to the stimulus.

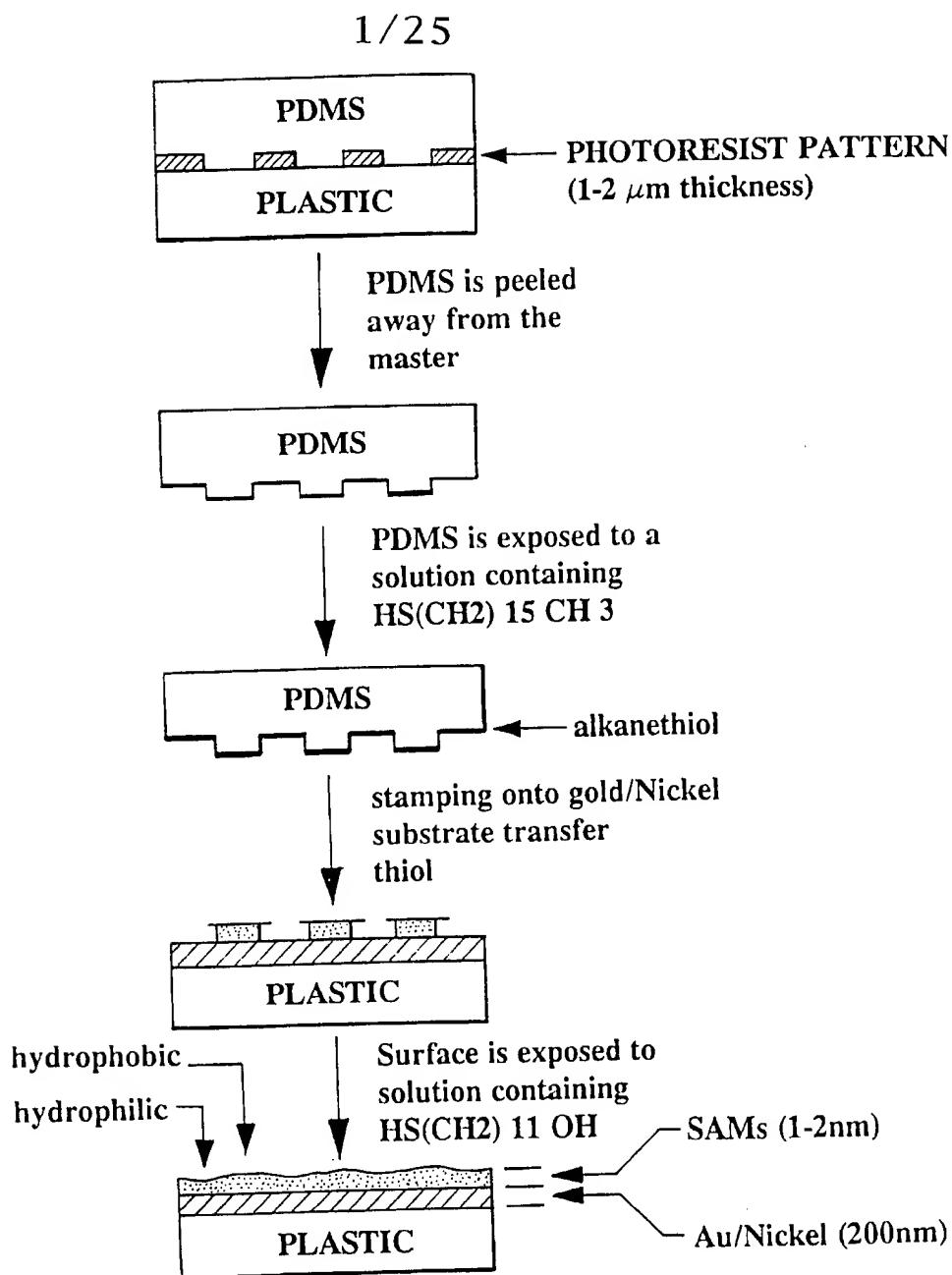


FIGURE 1

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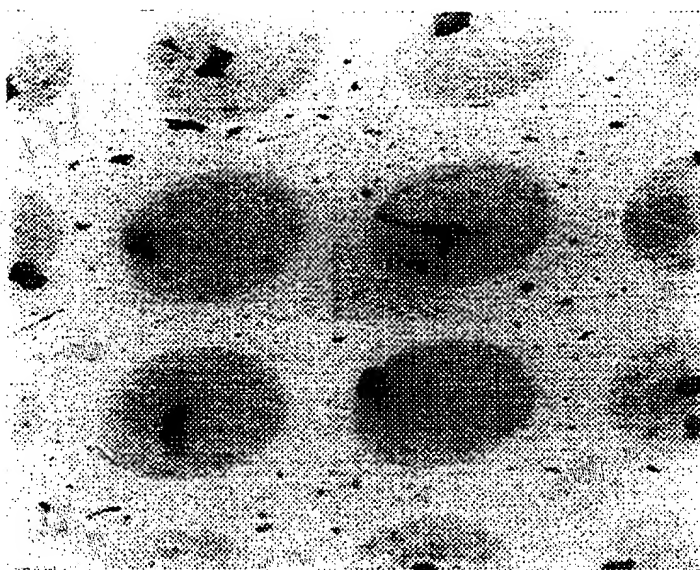


FIGURE 2

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FIGURE 3A

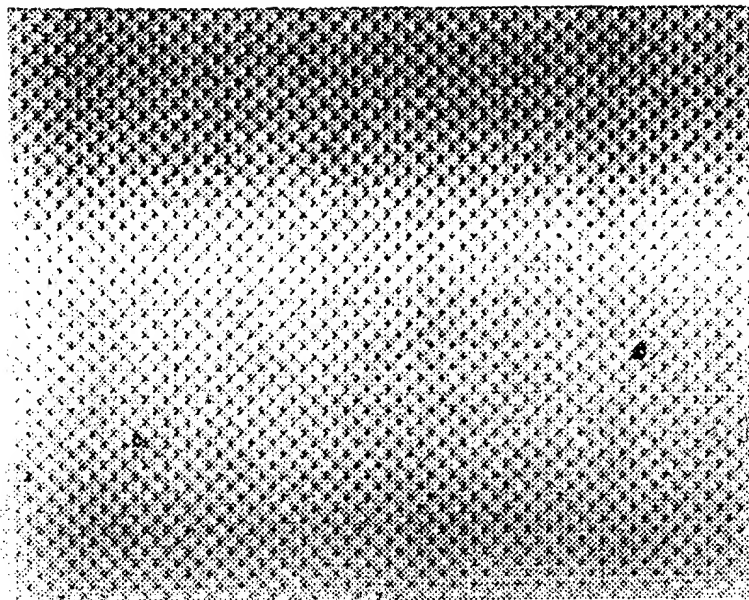
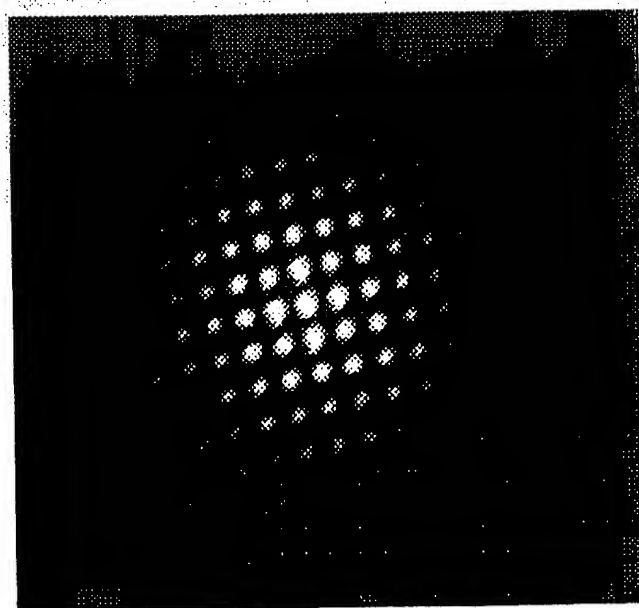


FIGURE 3B



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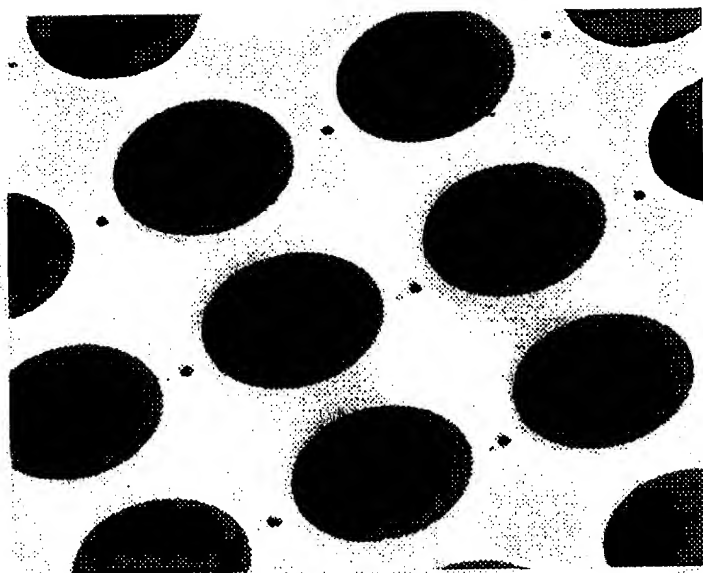


FIGURE 4

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Hydrophilic SAM Circle

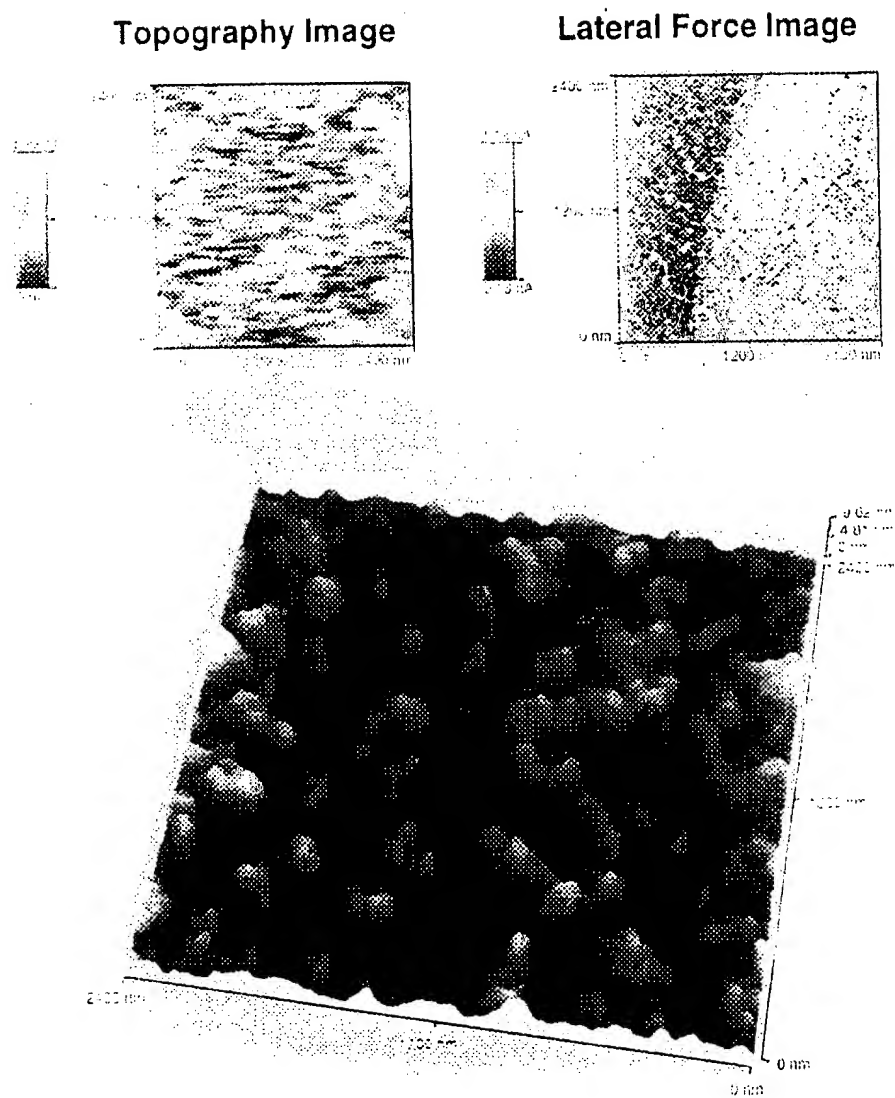


FIGURE 5

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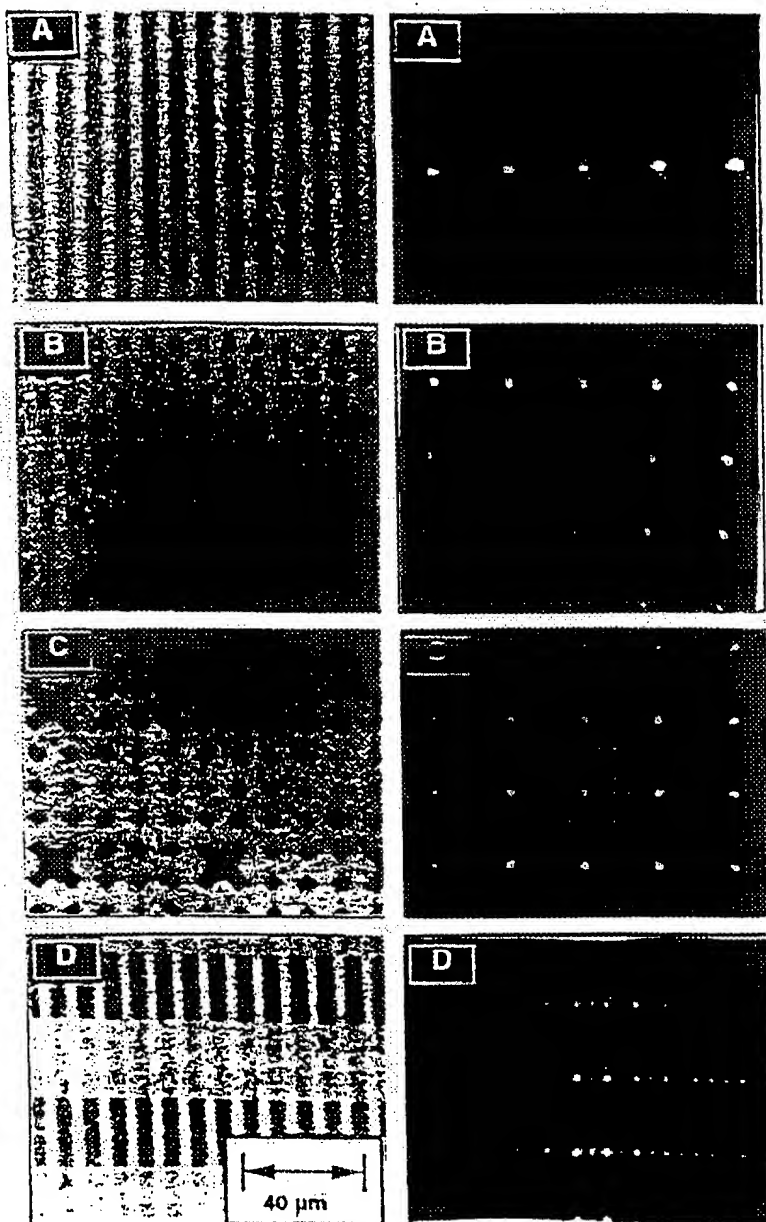


FIGURE 6

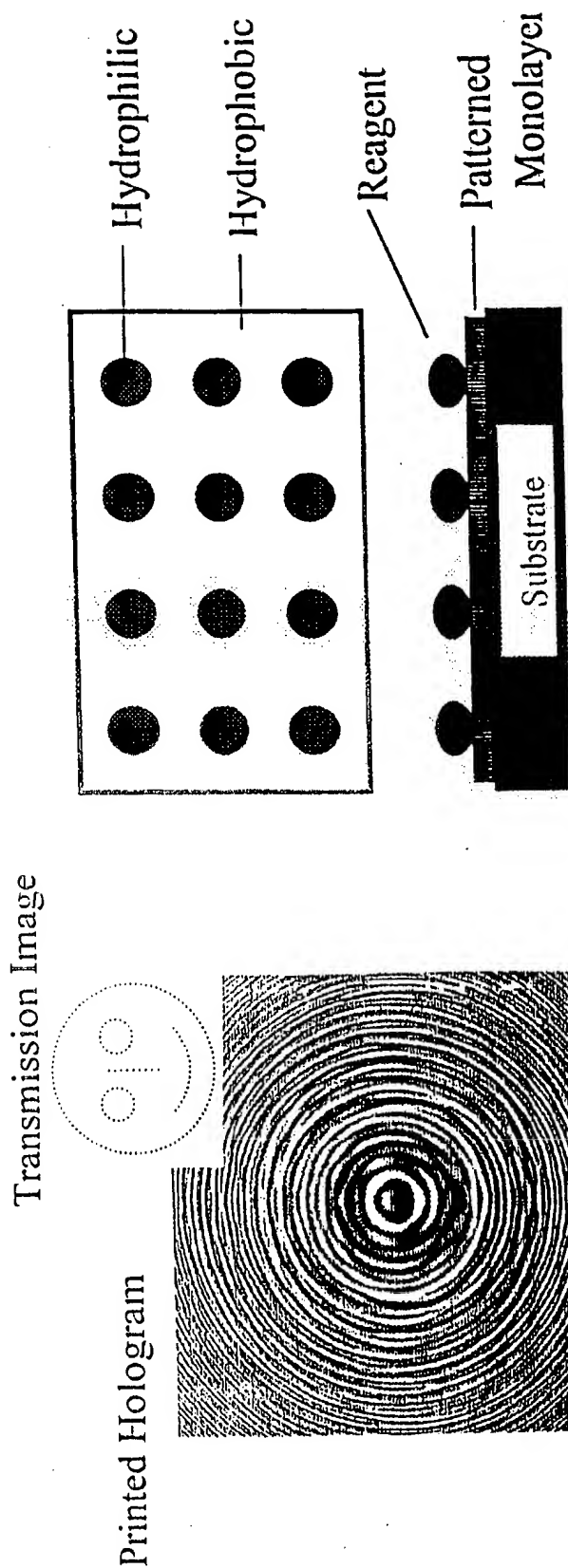
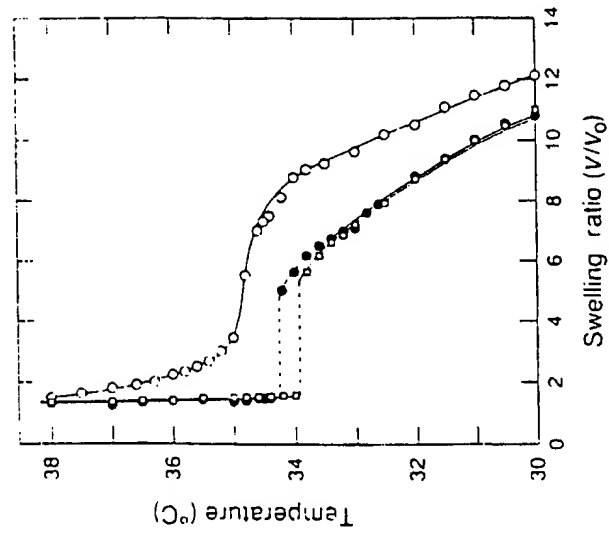


FIGURE 7

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Temp Induced Transition



Gel Phase Transition

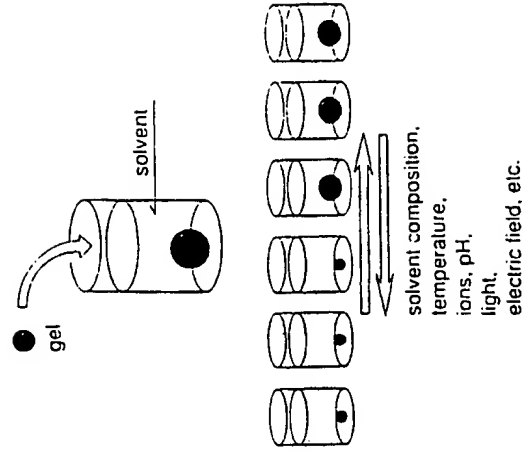
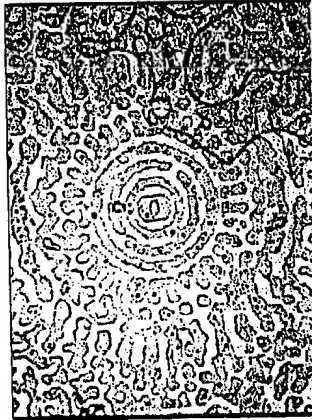


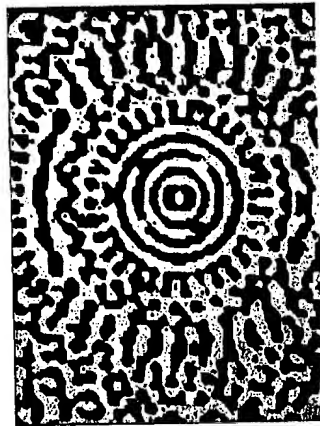
FIGURE 8

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Gel on metallized MYLAR

FIGURE 9B



High resolution printer film output

FIGURE 9A

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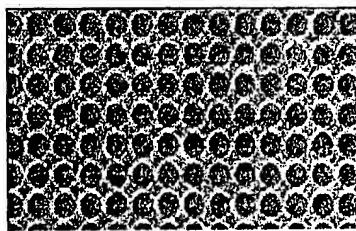


FIGURE 10

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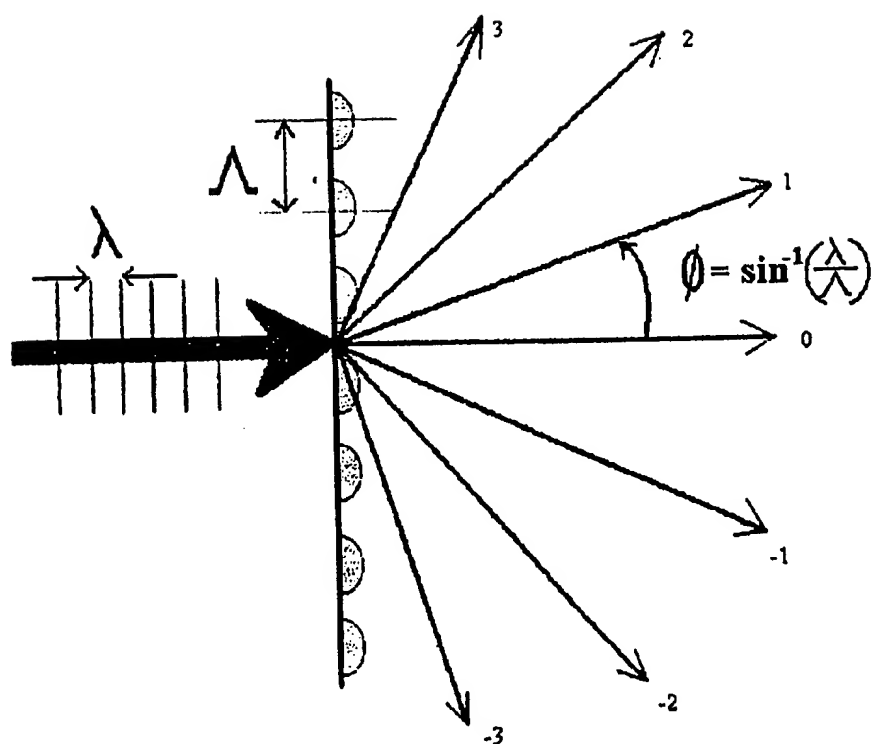


FIGURE 11

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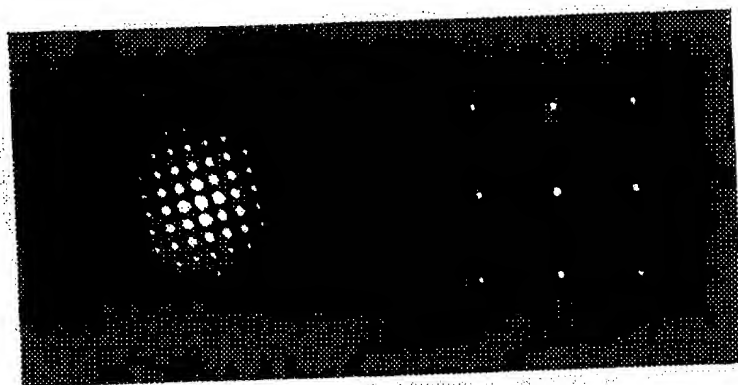


FIGURE 12

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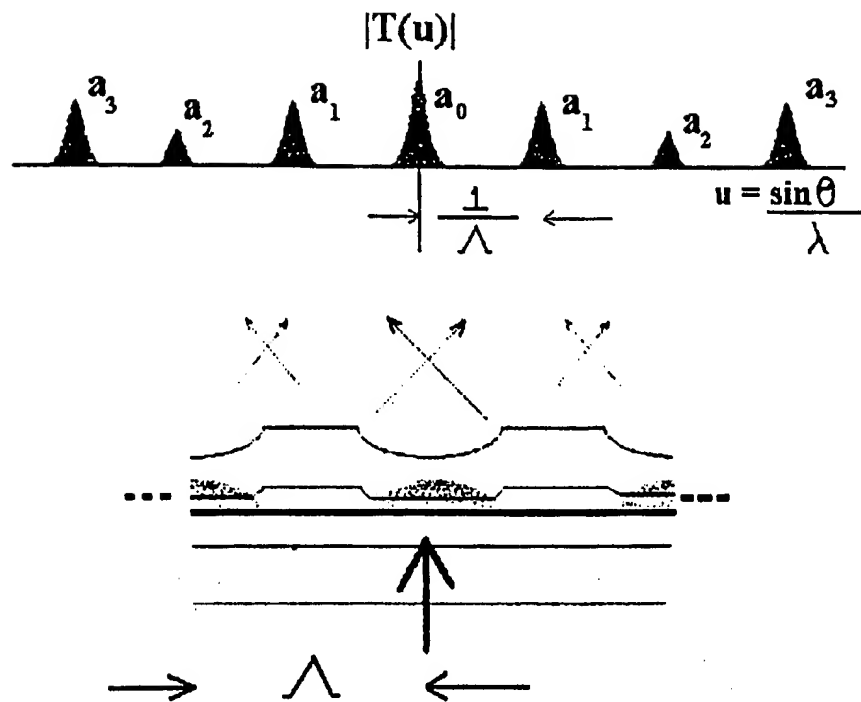


FIGURE 13

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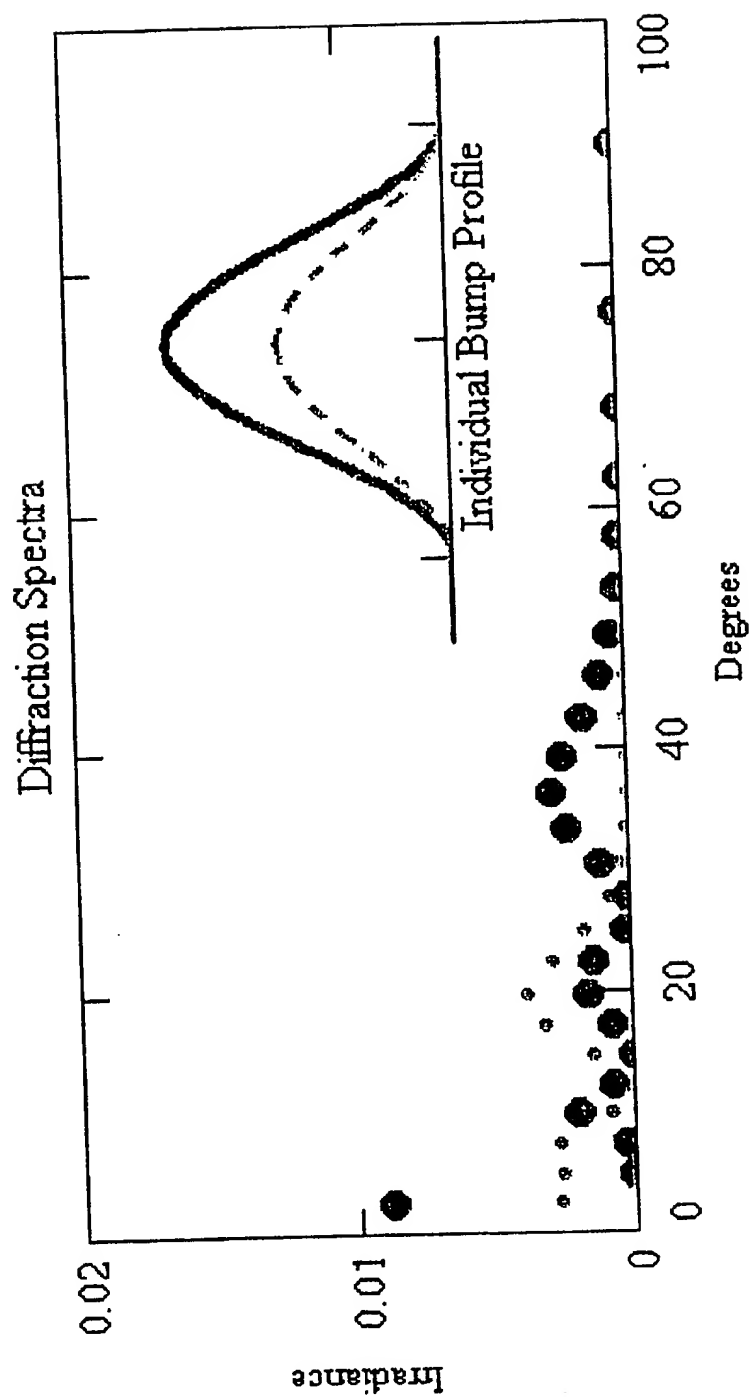


FIGURE 14

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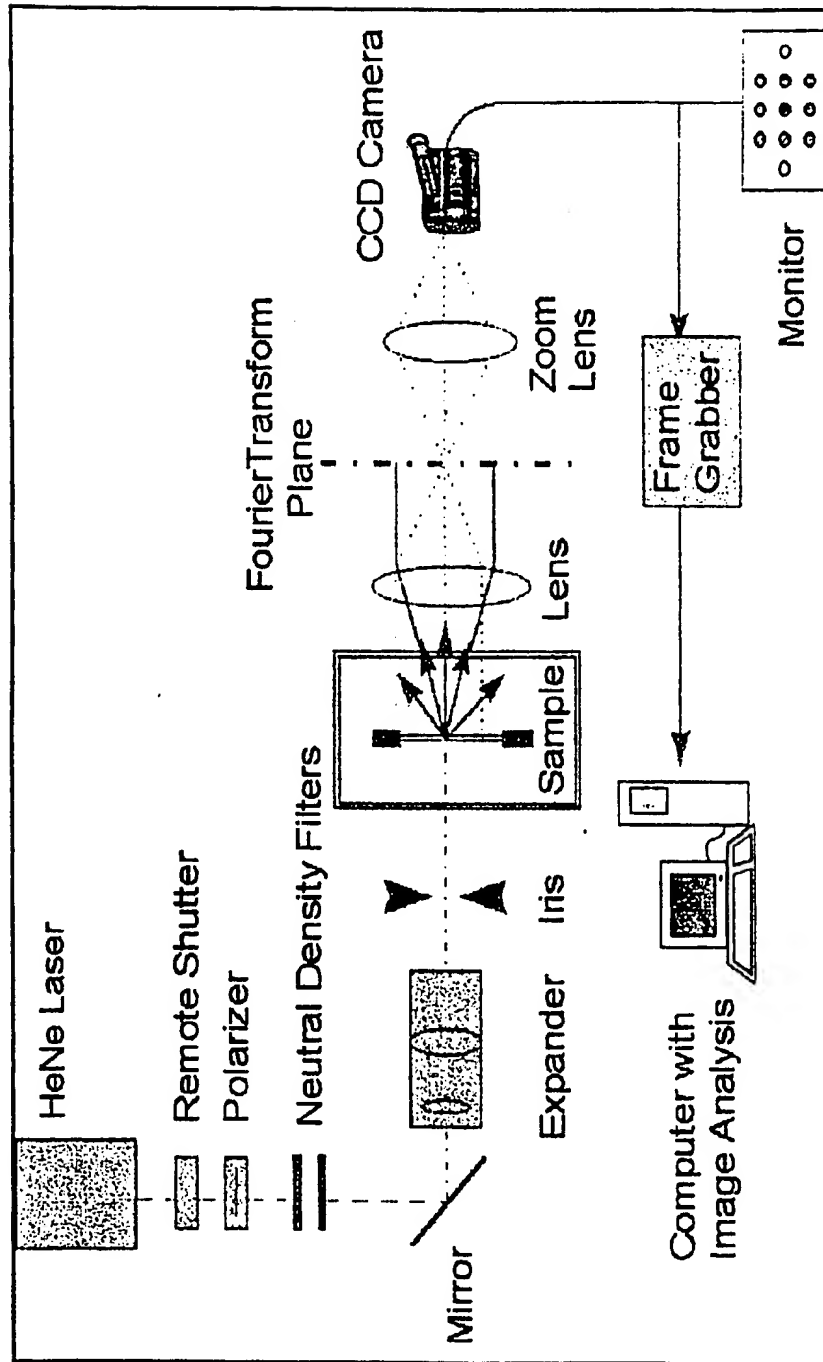


FIGURE 15

SUBSTITUTE SHEET (RULE 26)

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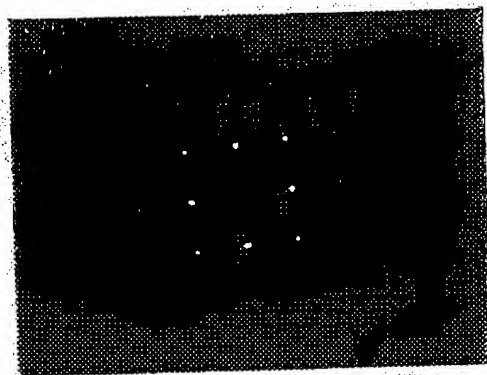


FIGURE 16

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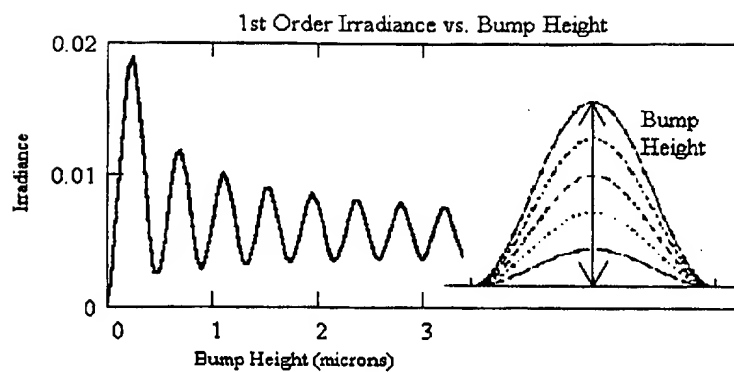


FIGURE 17

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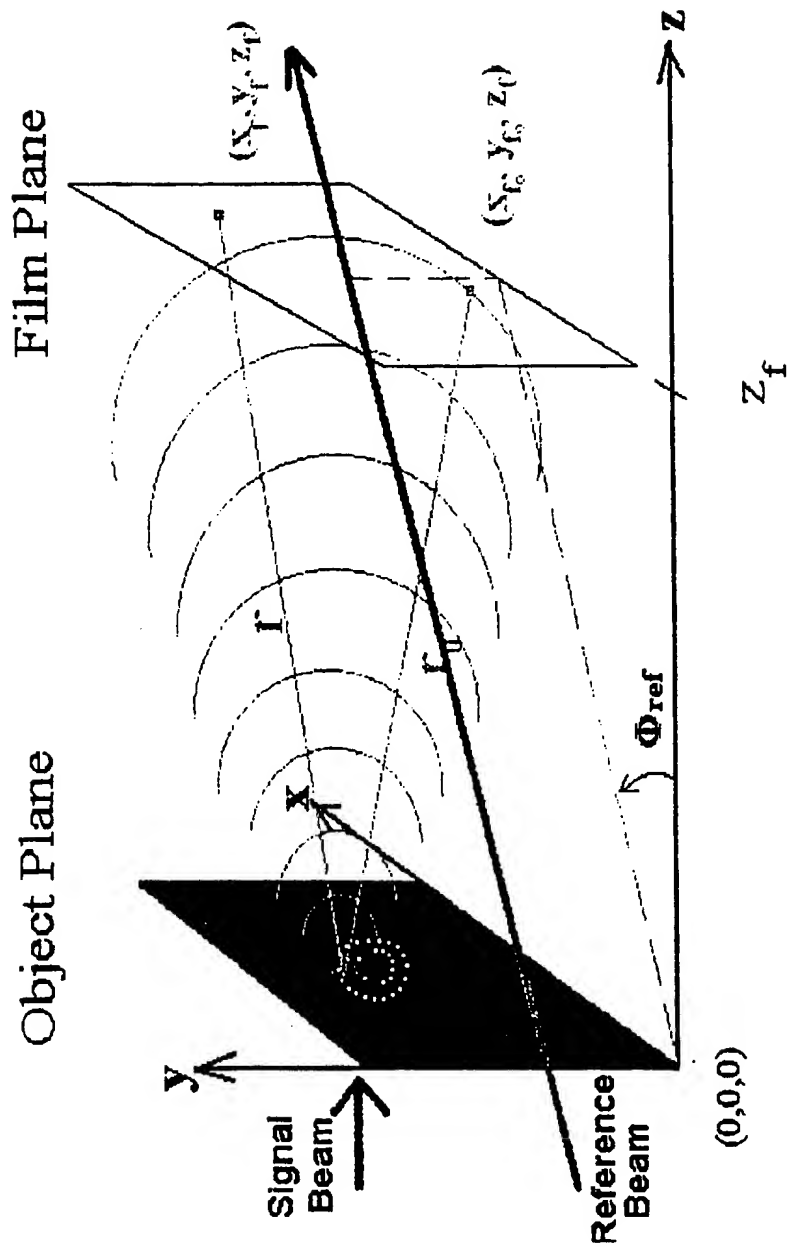


FIGURE 18

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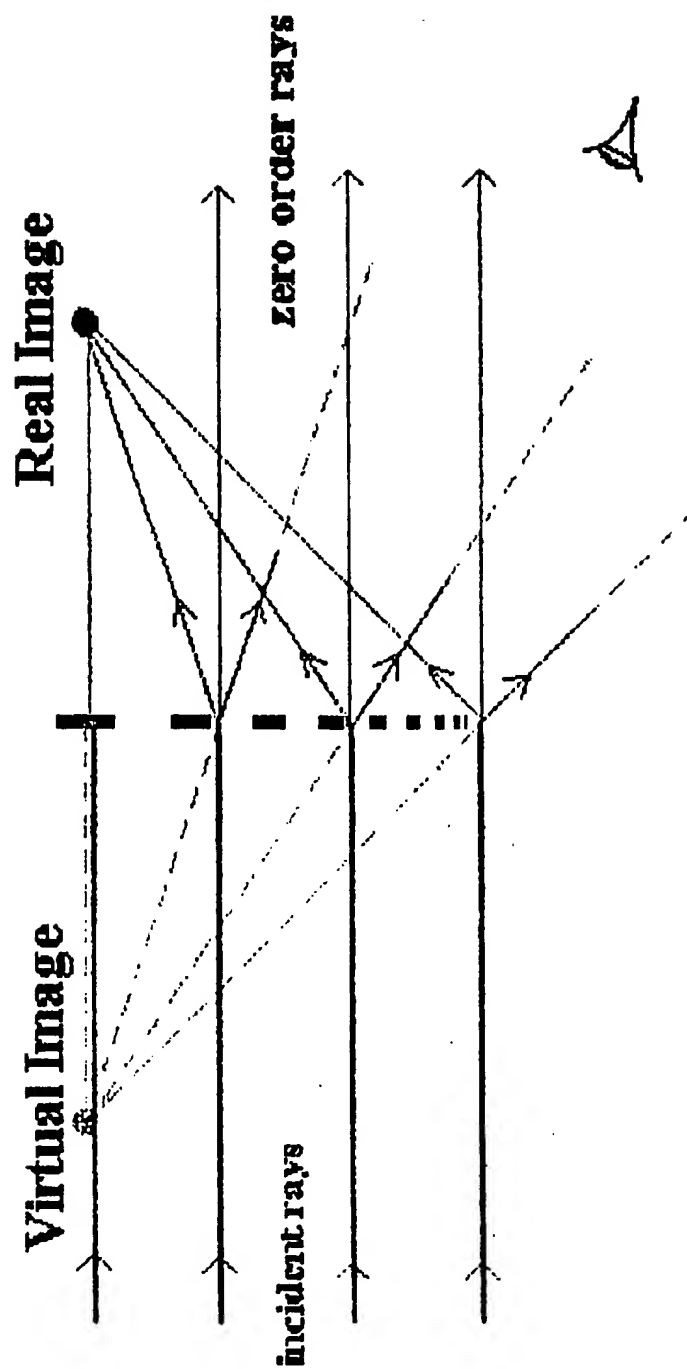


Figure 19

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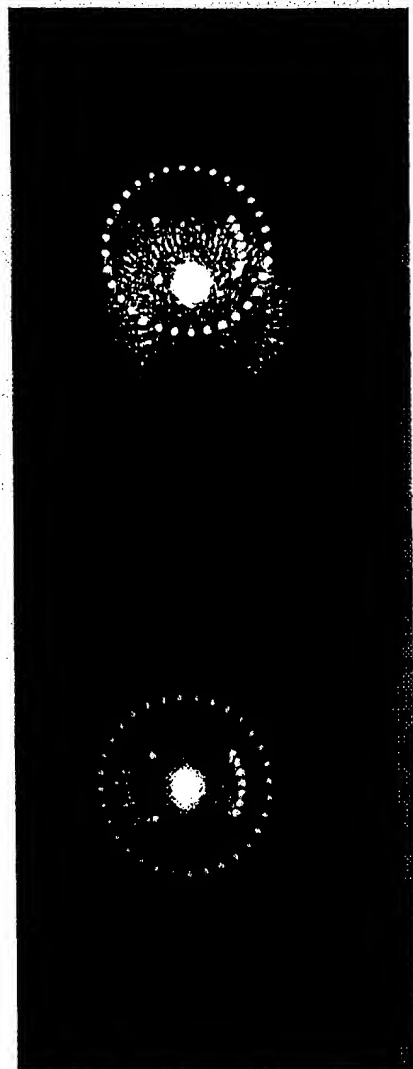


FIGURE 20

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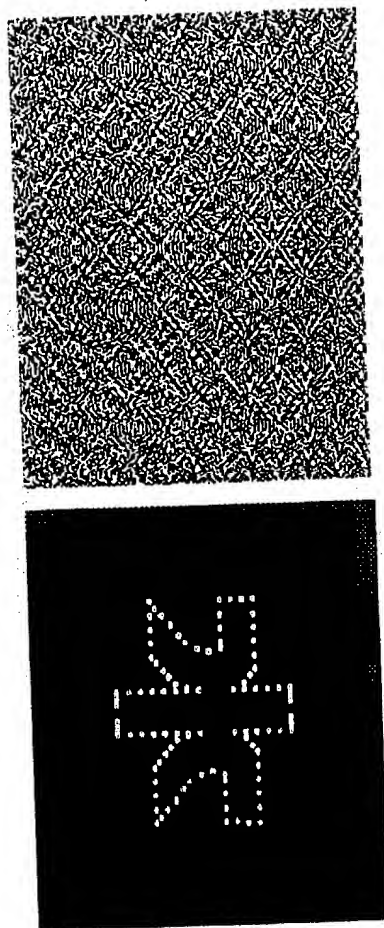


FIGURE 21

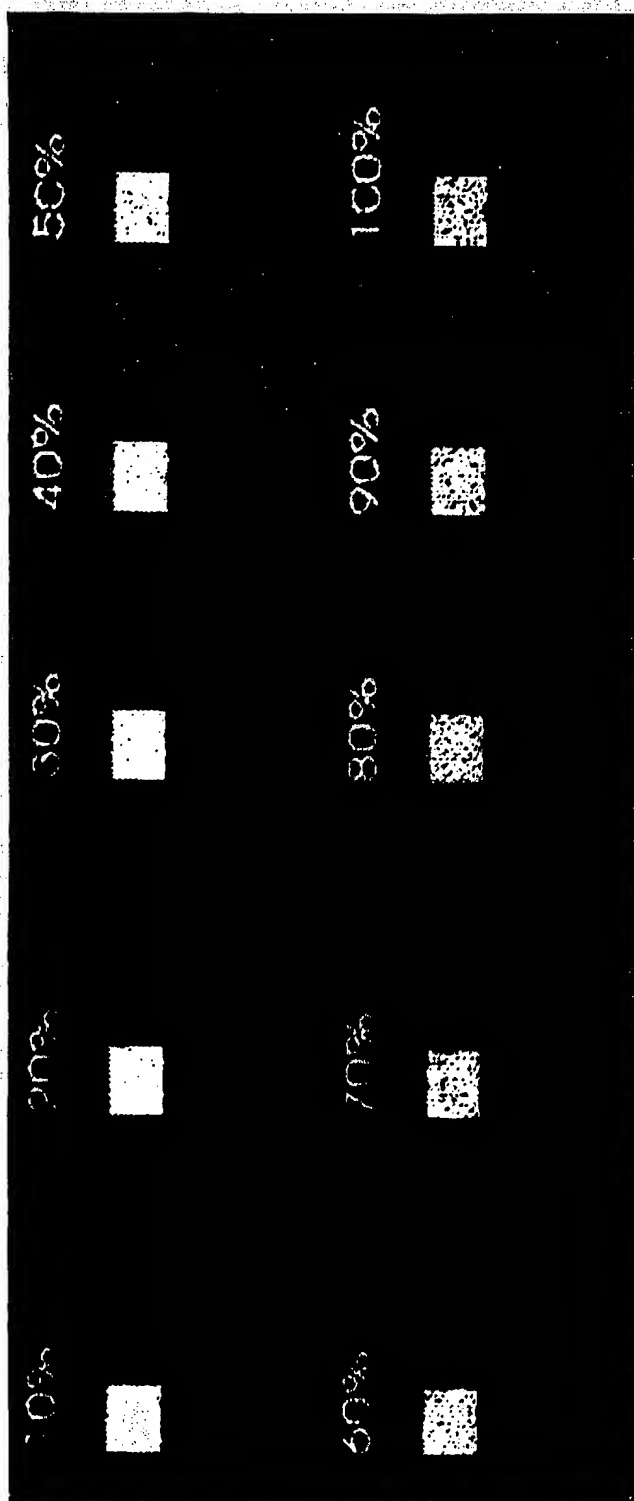


Figure 22A

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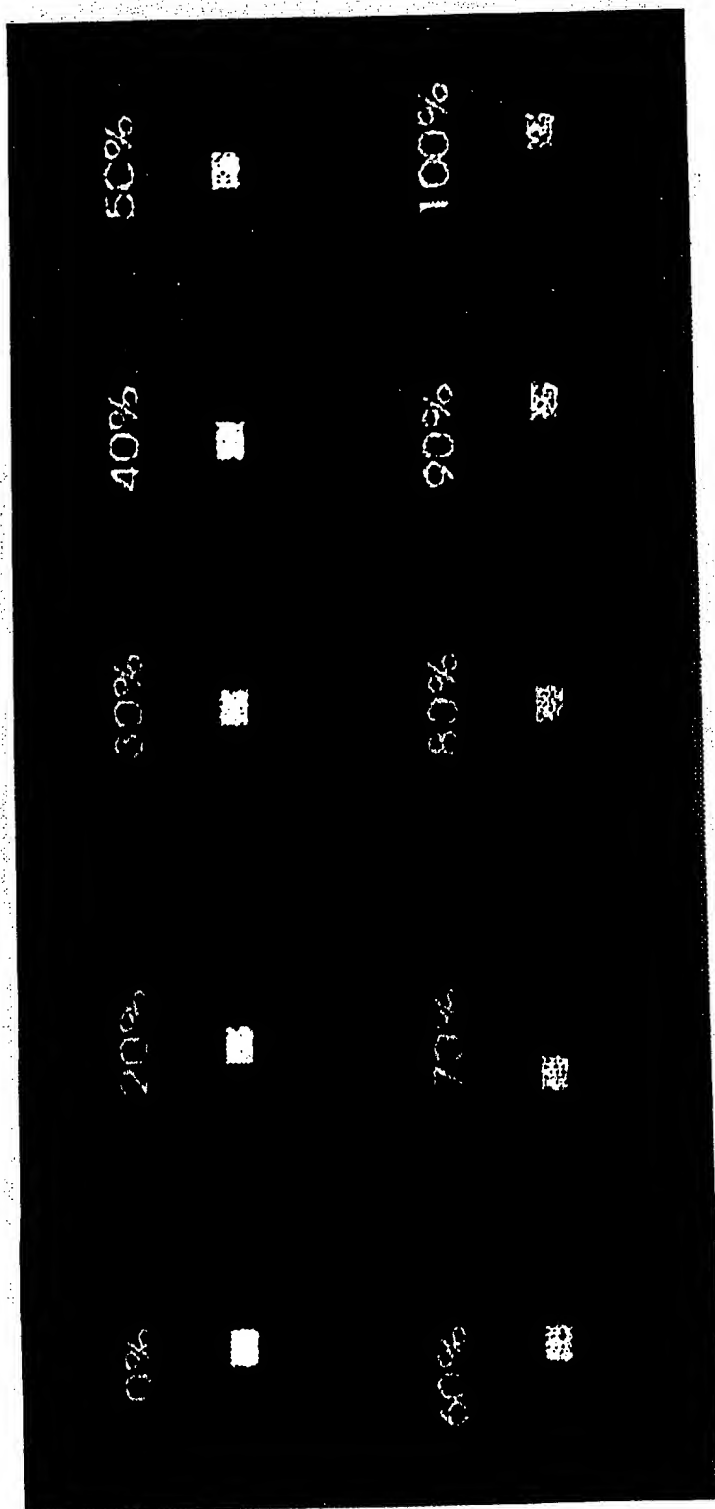


Figure 22B

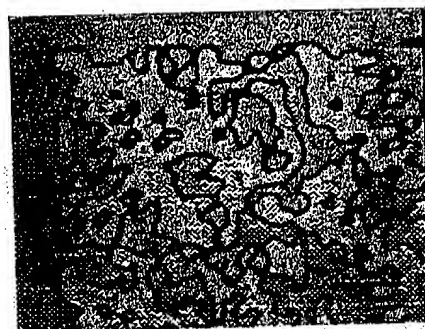
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FIGURE 23A

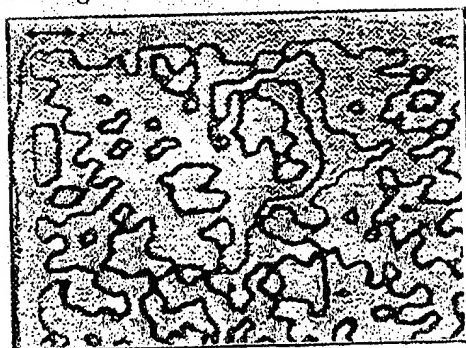


A. High-Resolution Printer film

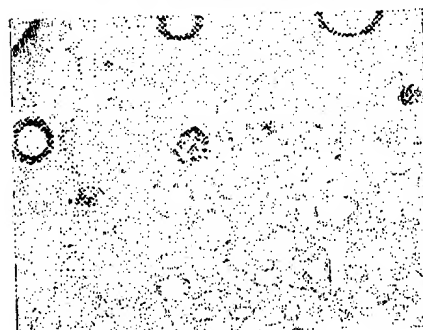
FIGURE 23B



B. Photoresist master on gold film



C. Elastomeric Stamp



D. Gel on metallized MYLAR

FIGURE 23C

FIGURE 23D

25/25

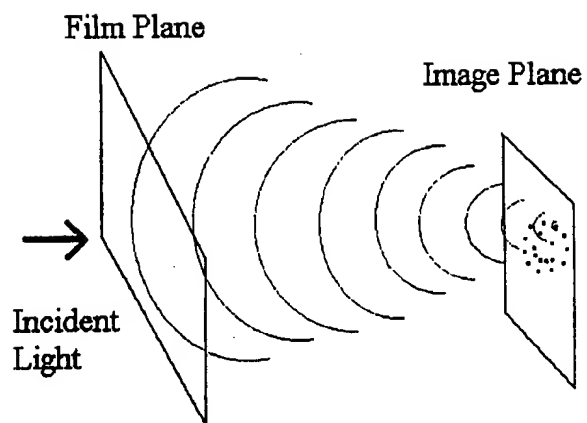


FIGURE 24

INTERNATIONAL SEARCH REPORT

In tional Application No
PCT/US 98/05400

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 G01N33/543 G03F7/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 G01N G03F B41M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P, X	WO 98 10334 A (KIMBERLY CLARK CO) 12 March 1998 see the whole document see page 3, paragraph 1 see page 4, line 3 ---	1-44
X	EP 0 596 421 A (HOFFMANN LA ROCHE) 11 May 1994 see claims see column 3, line 18 - column 4, line 21 see column 8, line 35 - column 9, line 19 see column 12, line 22 - column 13, line 37 ---	1-44
Y	---	1-44
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☒ Further documents are listed in the continuation of box C.

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Date of the actual completion of the international search

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 5 514 501 A (TARLOV MICHAEL J) 7 May 1996 see claims 1,8,12-15 see column 3, line 11 - line 21 see column 4, line 17 - line 67 see column 5, line 44 - line 60 ---	1-44
X	US 5 436 161 A (BERGSTROEM JAN ET AL) 25 July 1995 see claims Y see column 4, line 20 - column 6, line 19 ---	1-44
X	US 5 242 828 A (BERGSTROEM JAN ET AL) 7 September 1993 see claims Y see column 4, line 14 - column 5, line 2 see column 5, line 54 - column 6, line 35 ---	1-44
Y	PATENT ABSTRACTS OF JAPAN vol. 017, no. 499 (C-1109), 9 September 1993 & JP 05 132640 A (RICOH CO LTD), 28 May 1993, see abstract ---	1-10, 23-32
Y	PATENT ABSTRACTS OF JAPAN vol. 014, no. 286 (P-1064), 20 June 1990 & JP 02 085755 A (TEIJIN LTD), 27 March 1990, see abstract -----	1-10, 23-32

INTERNATIONAL SEARCH REPORT

Information on patent family members

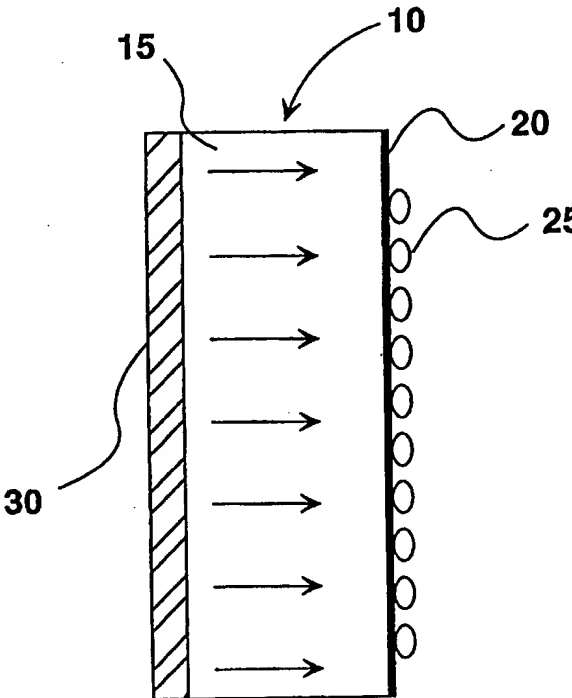
International Application No

PCT/US 98/05400

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 9810334 A	12-03-1998	AU 4181397 A	26-03-1998
EP 0596421 A	11-05-1994	CA 2108705 A	07-05-1994
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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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(21) International Application Number: PCT/US98/26759 (22) International Filing Date: 16 December 1998 (16.12.98) (30) Priority Data: 08/991,644 16 December 1997 (16.12.97) US (71) Applicant: KIMBERLY-CLARK WORLDWIDE, INC. [US/US]; 401 North Lake Street, Neenah, WI 54956 (US). (72) Inventors: EVERHART, Dennis, S.; 230 Hereford Road, Alpharetta, GA 30201 (US). JONES, Mark, L.; 823 Lake Avenue, Atlanta, GA 30307 (US). KAYLOR, Rosann, Marie; 7480 Williamsburg Drive, Cumming, GA 30131 (US). (74) Agents: GREEN, Theodore, M.; Jones & Askew, LLP, 2400 Monarch Tower, 3424 Peachtree Road, N.E., Atlanta, GA 30326 (US) et al.		(81) Designated States: AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG). Published <i>With international search report.</i> <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>
(54) Title: OPTICAL DIFFRACTION BIOSENSOR (57) Abstract <p>The present invention provides an inexpensive and sensitive device and method for detecting and quantifying analytes present in a medium. The device comprises a metalized film (20) upon which is printed a specific, pre-determined pattern of analyte-specific receptors (25). Upon attachment of a target analyte to select areas of the plastic film upon which the receptor is printed, diffraction of transmitted and/or reflected light occurs via the physical dimensions and defined, precise placement of the analyte. A diffraction image is produced which can be easily seen with the eye or, optionally, with a sensing device.</p> 		

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OPTICAL DIFFRACTION BIOSENSOR

TECHNICAL FIELD

15 The present invention is in the field of analyte sensors and, more specifically the present invention is in the field of microcontact printing binders on metal films to produce optical diffraction biosensors.

BACKGROUND OF THE INVENTION

20 Microcontact printing is a technique for forming patterns of organic monolayers with micron and submicron lateral dimensions. It offers experimental simplicity and flexibility in forming certain types of patterns. In the prior art, microcontact printing was used with self-assembled monolayers of long-chain alkanethiolates to form organic structures on gold and other metals. These patterns acted as nanometer resists by protecting the supporting metal from corrosion by appropriately formulated etchants, or, 25 allowed for the selective placement of fluids on hydrophilic regions of the pattern. In general, patterns of self-assembled monolayers having dimensions that can be less than 1 micron are formed by using the alkanethiol as an "ink", and by printing them on the metal support using an elastomeric "stamp". The stamp is fabricated by molding a silicone elastomer using a master prepared by optical or X-ray microlithography or by other techniques. (See U.S. Patent Application Serial Nos. 08/654,993; 08/769,594; 30 08/821,464; 08/707,456 and 08/768,449 which are incorporated herein in their entirety by reference)

35 Microcontact printing brings to microfabrication a number of new capabilities. Microcontact printing makes it possible to form patterns that are distinguished only by their constituent functional groups; this capability permits the control of surface properties such as interfacial free energies with great precision. In the prior art microcontact printing relies on molecular self-assembly. Using self-assembling monolayers, a system is generated that is (at least locally) close to a thermodynamic minimum and is intrinsically defect-rejecting and self-healing. Simple procedures, with minimal protection against surface contamination 40 by adsorbed materials or by particles, can lead to surprisingly low levels of defects in the

final structures. The procedure using self-assembling monolayers can be conducted at atmospheric pressure, in an unprotected laboratory atmosphere. Thus, microcontact printing that uses self-assembling monolayers is useful in laboratories that do not have routine access to the equipment normally used in microfabrication, or for which the capital cost of equipment is a serious concern. The patterned self-assembled monolayers can be designed to act as resists with a number of wet-chemical etchants.

Also in the prior art, a gold film 5 to 2000 nanometers thick is typically supported on a titanium-primed Si/SiO₂ wafer or glass sheet. The titanium serves as an adhesion promoter between gold and the support. However, the silicon wafer is rigid, brittle, and cannot transmit light. These silicon wafers are also not suitable for a large-scale, continuous printing process, such as in letterpress, gravure, offset, and screen printing (see Printing Fundamentals, A. Glassman, Ed. (Tappi Press Atlanta, GA 1981); Encyclopedia Britannica, vol. 26, pp. 76-92, 110-111 (Encyclopedia Britannica, Inc. 1991)). In addition, silicon must be treated in a separate step with an adhesion promoter such as Cr or Ti, or Au will not adequately adhere, preventing formation of a stable and well-ordered monolayer. Finally, silicon is opaque to visible light, so any diffraction pattern obtained must be created with reflected, not transmitted light.

What is needed is an easy, efficient and simple method of contact printing a patterned receptor on an optically transparent, flexible substrate, that is amenable to continuous processing and does not use self-assembling monolayers. Such a method and the device resulting from such a method is simpler, not restricted to the limitations of self-assembling monolayers and is easier to manufacture.

SUMMARY OF THE INVENTION

The present invention provides an inexpensive and sensitive device and method for detecting and quantifying analytes present in a medium. The device comprises a metalized film upon which is printed a specific predetermined pattern of analyte-specific receptor. The present invention does not utilize self-assembling monolayers but is more general in that any receptor which can be chemically coupled to a surface can be used. Upon attachment of a target analyte which is capable of scattering light to select areas of the plastic film upon which the receptor is printed, diffraction of transmitted and/or reflected light occurs via the physical dimensions, refractive index and defined, precise placement of the analyte. In the case where an analyte does not scatter visible light because the analyte is too small or does not have an appreciable refractive index difference compared to the surrounding medium, the attachment of polymer beads coupled with the analyte to receptors is another method of producing diffraction of light. A diffraction image is

produced which can be easily seen with the eye or, optionally, with a sensing device. The present invention is a biosensor comprising a polymer film coated with metal and a receptor layer printed onto the polymer film wherein the receptor layer has a receptive material thereon that specifically binds an analyte.

5 The present invention utilizes methods of contact printing of patterned monolayers utilizing derivatives of binders for microorganisms. One example of such a derivative is a thiol. The desired binders can be thiolated antibodies or antibody fragments, proteins, nucleic acids, sugars, carbohydrates, or any other functionality capable of binding an analyte. The derivatives are chemisorbed to metal surfaces such as metalized polymer
10 films.

 Patterned monolayers allow for the controlled placement of analytes thereon via the patterns of analyte-specific receptors. The biosensing devices of the present invention produced thereby are used by first exposing the biosensing device to a medium that contains the analyte of choice and then, after an appropriate incubation period, transmitting
15 light, such as from a laser or a point light source, through the film. If the analyte is present in the medium and is bound to the receptors on the patterned monolayer, the light is diffracted in such a way as to produce a visible or near infrared image. In other words, the patterned monolayers with the analyte bound thereto can produce optical diffraction patterns which differ depending on the reaction of the receptors on the monolayer with the
20 analyte of interest. The light can be in the visible spectrum, and be either reflected from the film, or transmitted through it, and the analyte can be any compound or particle reacting with the monolayer. The light can be a white light or monochromatic electromagnetic radiation in preferably the visible region. The present invention also provides a flexible support for a monolayer on gold or other suitable metal or metal alloy.

25 The present invention includes a support for a thin layer of gold or other suitable material which does not require an adhesion promoter for the formation of a well-ordered monolayer or thin layer of binder. The present invention also provides a support for a layer of gold or other material which is suitable for continuous printing, rather than batch, fabrication. In addition, the present invention provides a low-cost, disposable biosensor
30 which can be mass produced. The biosensors of the present invention can be produced as a single test for detecting an analyte or it can be formatted as a multiple test device. The uses for the biosensors of the present invention include, but are not limited to, detection of chemical or biological contamination in garments, such as diapers, generally the detection of contamination by microorganisms in prepacked foods such as fruit juices or other
35 beverages and the use of the biosensors of the present invention in health diagnostic applications such as diagnostic kits for the detection of antigens, microorganisms, and blood constituents.

In another embodiment of the present invention, nutrients for a specific class of microorganisms can be incorporated into the receptor monolayer. In this way, very low concentrations of microorganisms can be detected by first contacting the biosensor of the present invention with the nutrients incorporated therein and then incubating the biosensor under conditions appropriate for the growth of the bound microorganism. The microorganism is allowed to grow until there are enough organisms to form a diffraction pattern.

The present invention can also be used on contact lenses, eyeglasses, window panes, pharmaceutical vials, solvent containers, water bottles, bandaids, and the like to detect contamination.

These and other features and advantages of the present invention will become apparent after a review of the following detailed description of the disclosed embodiments.

BRIEF DESCRIPTION OF THE FIGURES

Figure 1 is a schematic representation of a metal plated MYLAR film with a nutrient backing.

Figure 2 shows a biosensor capable of simultaneously measuring several different analytes in a medium.

Figure 3 is a schematic of contact printing of receptors according to the present invention.

Figure 4 is an enzyme-linked immunosorbent assay (ELISA) of the surface printed with a thiolated antibody binder.

Figure 5 is a photograph showing polystyrene surrogate particles coated with antigen after attachment to the printed antibody.

Figure 6 is a diffraction pattern produced from the sample described in Figure 4.

Figure 7 is an optical photomicrograph of *Candida albicans* attached to a patterned antibody receptor.

Figure 8 is a diffraction pattern caused by the binding of *Candida albicans* to the patterned receptor.

DETAILED DESCRIPTION

The present invention features improved biosensing devices, and methods for using such biosensing devices, for detecting and quantifying the presence or amount of an analyte of interest within a medium. The analytes that can be detected by the present invention include, but are not limited to, microorganisms such as bacteria, yeasts, fungi and viruses. In contrast to prior devices, those of the present invention allow detection of extremely small quantities of analyte in a medium in a rapid assay lasting only a few

minutes. In addition, other than a light source, no signaling or associated electronic components are required in the biosensing devices of the present invention.

5 The present invention comprises micro-contact printing of analyte-specific receptors (thiolated binders) onto metalized plastic film which allows for the development of single use, disposable biosensors based on light diffraction to indicate the presence of the analyte. Upon attachment of a target analyte to select areas of the plastic film which contain the receptor, diffraction of transmitted and/or reflected light occurs via the physical dimensions and defined, precise placement of the analyte. For example, yeast, fungi or bacterium are large enough to act as diffraction elements for visible light when placed in organized
10 patterns on a surface.

In addition to producing a simple diffraction image, patterns of analytes can be such as to allow for the development of a holographic sensing image and/or a change in visible color. Thus, the appearance of a hologram or a change in an existing hologram will indicate a positive response. The pattern made by the diffraction of the transmitted light can be any
15 shape including, but not limited to, the transformation of a pattern from one pattern to another upon binding of the analyte to the receptive material. In particularly preferred embodiments, the diffraction pattern becomes discernible in less than one hour after contact of the analyte with the biosensing device of the present invention.

The diffraction grating which produces the diffraction of light upon interaction with
20 the analyte must have a minimum periodicity of about $1/2$ the wavelength and real or imaginary a refractive index different from that of the surrounding medium. Very small analytes, such as viruses or molecules, can be detected indirectly by using a larger particle that is specific for the small analyte. One embodiment in which the small analyte can be detected comprises coating the particle, such as a latex bead or polystyrene bead, with a
25 receptive material, such as an antibody, that specifically binds to the analyte of interest. Particles that can be used in the present invention include, but are not limited to, glass, cellulose, synthetic polymers or plastics, latex, polystyrene, polycarbonate, proteins, bacterial or fungal cells and the like. The particles are desirably spherical in shape, but the structural and spatial configuration of the particles is not critical to the present invention.
30 For instance, the particles could be slivers, ellipsoids, cubes, random shape and the like. A desirable particle size ranges from a diameter of approximately $0.2\text{ }\mu\text{m}$ to $50\text{ }\mu\text{m}$, desirably between approximately $0.4\text{ }\mu\text{m}$ to $1\text{ }\mu\text{m}$. The composition of the particle is not critical to the present invention.

35 It is to be understood that the optimal particle size is a function of the refractive index of the particle and the refractive index of the surrounding medium. A method of analyzing the optimal particle size for use in the present invention with a transmission image is by employing the equation;

$$t_{\text{opt}} = \lambda/2(n_2 - n_1)$$

wherein t_{opt} = optimum height of the particle

λ = wavelength of incoming light

n_2 = refractive index of particle

5

n_1 = refractive index of surrounding medium

For a reflection image, the optimum height of the particle is determined by the above equation divided by two.

10

The monolayer on the metalized film contains a receptive material or binder, such as an antibody, that will specifically bind to an epitope on the analyte that is different from the epitope used in the binding to the particle. Thus, for detecting a medium with a small analyte, such as viral particles, the medium is first exposed to the latex particles to which the viral particles are bound. Then, the latex particles are optionally washed and exposed to the metalized film with the monolayers containing the virus specific antibodies. The antibodies then bind to the viral particles on the latex bead thereby immobilizing the latex beads in the same pattern as the monolayers on the film. Because the bound latex beads will cause diffraction of the visible light, a diffraction pattern is formed, indicating the presence of the viral particle in the liquid. Other combinations using particles are well known to those of ordinary skill in the art.

15

20

In another embodiment of the present invention, receptors, such as antibodies are attached to the metal layer as described herein. The antibodies are then exposed to an environment that contains analytes that bind to the receptor. After the analyte has bound to the receptor, a second receptor is added that recognizes the metal bound conjugate. To this second receptor is bound an enzyme or inorganic substance that will cause a precipitation of a solid substance when the appropriate reagent or reagents are added. For example, an enzyme that can cause a precipitate to form is peroxidase in the presence of tetramethylbenzidine (See Example 3 herein). Another example, is the use of colloidal gold in the presence of a silver halide. Elemental silver will precipitate on the patterned receptor layer thereby producing the diffraction pattern.

25

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The analytes that are contemplated as being detected using the present invention include, but are not limited to, bacteria; yeasts; fungi; viruses; rheumatoid factor; antibodies, including, but not limited to IgG, IgM, IgA and IgE antibodies; carcinoembryonic antigen; streptococcus Group A antigen; viral antigens; antigens associated with autoimmune disease, allergens, tumor antigens; streptococcus Group B antigen, HIV I or HIV II antigen; or host response (antibodies) to these and other viruses; antigens specific to RSV or host response (antibodies) to the virus; antigen; enzyme;

35

hormone; polysaccharide; protein; lipid; carbohydrate; drug or nucleic acid; *Salmonella species*; *Candida species*, including, but not limited to *Candida albicans* and *Candida tropicalis*; *Neisseria meningitidis* groups A, B, C, Y and W sub 135, *Streptococcus pneumoniae*, *E. coli*, *Haemophilus influenza* type B; an antigen derived from microorganisms; a hapten; a drug of abuse; a therapeutic drug; an environmental agent; and antigens specific to Hepatitis.

In another embodiment of the present invention, nutrients for a specific class of microorganism can be incorporated into the monolayer. In this way, very low concentrations of microorganisms can be detected by first contacting the biosensor of the present invention with the nutrients incorporated therein and then incubating the biosensor under conditions appropriate for the growth of the bound microorganism. In one embodiment shown in Figure 1, the MYLAR film 15 has a nutrient backing 30 that is in contact with the back of the MYLAR film 15. The opposite side of the MYLAR film 15 has a metal film 20 thereon. The metal film 20 is preferably gold. Attached to the metal film 20 are receptors 25 that are specific for a microorganism. In use, the nutrient diffuses slowly through the MYLAR film. When a microorganism is attached to receptor 25, the bound microorganism consumes the nutrient and grows. As the microorganism grows, it diffracts impinging light thereby forming a diffraction pattern. Thus, in this embodiment, if the diffraction pattern forms, it is because the bound microorganism grew. Of course, in some cases, the microorganism can multiply enough to form a diffraction pattern without the presence of a nutrient on the patterned monolayer.

A part of the present invention is a receptive material that can be microprinted on the metalized film and will specifically bind to the analyte of interest. Thus, the receptive material is defined as one part of a specific binding pair and includes, but is not limited to, antigen/ antibody, enzyme/substrate, oligonucleotide/DNA, chelator/metal, enzyme/inhibitor, bacteria/receptor, virus/receptor, hormone/receptor, DNA/RNA, or RNA/RNA, oligonucleotide /RNA, and binding of these species to any other species, as well as the interaction of these species with inorganic species.

The receptive material that is bound to the attachment layer is characterized by an ability to specifically bind the analyte or analytes of interest. The variety of materials that can be used as receptive material is limited only by the types of material which will combine selectively (with respect to any chosen sample) with a secondary partner. Subclasses of materials which fall in the overall class of receptive materials include toxins, antibodies, antibody fragments, antigens, hormone receptors, parasites, cells, haptens, metabolites, allergens, nucleic acids, nuclear materials, autoantibodies, blood proteins, cellular debris, enzymes, tissue proteins, enzyme substrates, coenzymes, neuron transmitters, viruses, viral particles, microorganisms, proteins, polysaccharides, chelators,

drugs, and any other member of a specific binding pair. This list only incorporates some of the many different materials that can be coated onto the attachment layer to produce a thin film assay system. Whatever the selected analyte of interest is, the receptive material is designed to bind specifically with the analyte of interest.

5 The matrix containing the analyte of interest may be a liquid, a solid, or a gas, and can include a bodily fluid such as mucous, saliva, urine, fecal material, tissue, marrow, cerebral spinal fluid, serum, plasma, whole blood, sputum, buffered solutions, extracted solutions, semen, vaginal secretions, pericardial, gastric, peritoneal, pleural, or other washes and the like. The analyte of interest may be an antigen, an antibody, an enzyme, a DNA fragment, an intact gene, a RNA fragment, a small molecule, a metal, a toxin, an environmental agent, a nucleic acid, a cytoplasm component, pili or flagella component, protein, polysaccharide, drug, or any other material, such as those listed in Table A. For example, receptive material for bacteria may specifically bind a surface membrane component, protein or lipid, a polysaccharide, a nucleic acid, or an enzyme. The analyte which is specific to the bacteria may be a polysaccharide, an enzyme, a nucleic acid, a membrane component, or an antibody produced by the host in response to the bacteria. The presence of the analyte may indicate an infectious disease (bacterial or viral), cancer or other metabolic disorder or condition. The presence of the analyte may be an indication of food poisoning or other toxic exposure. The analyte may indicate drug abuse or may monitor levels of therapeutic agents.

20 One of the most commonly encountered assay protocols for which this technology can be utilized is an immunoassay. However, the general considerations apply to nucleic acid probes, enzyme/substrate, and other ligand/receptor assay formats. For immunoassays, an antibody may serve as the receptive material or it may be the analyte of interest. The receptive material, for example an antibody or an antigen, must form a stable, dense, reactive layer on the attachment layer of the test device. If an antigen is to be detected and an antibody is the receptive material, the antibody must be specific to the antigen of interest; and the antibody (receptive material) must bind the antigen (analyte) with sufficient avidity that the antigen is retained at the test surface. In some cases, the analyte may not simply bind the receptive material, but may cause a detectable modification of the receptive material to occur. This interaction could cause an increase in mass at the test surface or a decrease in the amount of receptive material on the test surface. An example of the latter is the interaction of a degradative enzyme or material with a specific, immobilized substrate. In this case, one would see a diffraction pattern before interaction with the analyte of interest, but the diffraction pattern would disappear if the analyte were present. The specific mechanism through which binding, hybridization, or interaction of

the analyte with the receptive material occurs is not important to this invention, but may impact the reaction conditions used in the final assay protocol.

5 In general, the receptive material may be passively adhered to the attachment layer in a pattern that will produce a diffraction pattern. If required, the free functional groups introduced onto the test surface by the attachment layer may be used for covalent attachment of receptive material to the test surface. Chemistries available for attachment of receptive materials are well known to those skilled in the art.

10 A wide range of techniques can be used to adhere the receptive material to the attachment layer in a pattern that, when bound to the analyte of interest, forms a diffraction pattern when light is transmitted through attachment layer. Test surfaces may be coated with receptive material by application of solution in discrete arrays or patterns; spraying, ink jet, or other imprinting methods; or by spin coating from an appropriate solvent system. The technique selected should minimize the amount of receptive material required for coating a large number of test surfaces and maintain the stability/functionality of receptive material during application. The technique must also apply or adhere the receptive material to the attachment layer in a very uniform and reproducible fashion.

20 The receptor layer is formed from material selected from the group consisting of antigens, antibodies, oligonucleotides, chelators, enzymes, bacteria, bacterial pili, bacterial flagellar materials, nucleic acids, polysaccharides, lipids, proteins, carbohydrates, metals, viruses, hormones and receptors for said materials. In the preferred embodiments, the biosensing device is configured and arranged to provide a pattern detectable by eye in response to transmission of polychromatic light when the analyte of interest is sandwiched between the receptive material and a secondary binding reagent.

25 The medium in which the analyte may reside can be solid, gel-like, liquid or gas. For purposes of detecting an analyte in a body fluid, the fluid is selected from the group consisting of urine, serum, plasma, spinal fluid, sputum, whole blood, saliva, uro-genital secretions, fecal extracts, pericardial, gastric, peritoneal, pleural washes, vaginal secretions, and a throat swab; and the method optionally includes using a diffractometer to measure the appearance of the diffraction pattern. The most common gas that is contemplated as being used with the biosensing device of the present invention is air.

30 The biosensing device of the present invention utilizes methods of contact printing of patterned monolayers on metalized polymer films, desirably thermoplastic polymer films, the compositions produced thereby, and the use of these compositions. Patterned monolayers allow for the controlled placement of fluids thereon which can contain an analyte receptor. The term "patterned monolayers thereon" as used herein means the monolayers in any pattern on the metalized polymer films including a solid pattern.

35

When the film with the monolayers thereon is exposed to an analyte that is capable of reacting with the monolayer, the film will produce optical diffraction patterns which differ depending on the reaction of the monolayer with the analyte of interest. The liquid may be a high surface tension fluid such as water. The light can be in the visible spectrum, and be either reflected from the film, or transmitted through it, and the analyte can be any compound reacting with the monolayer.

In preferred embodiments, the method involves contacting the substrate with a test sample potentially containing the analyte under conditions in which the substrate causes a change in the refractive index of the monolayer. When light is transmitted through the metalized thermoplastic polymer with the monolayer, a visible pattern is formed and can be visualized by directing the light to a surface or by looking directly through the substrate.

In one embodiment, the present invention is contemplated in a dipstick form in which the micro-contact printed metalized film is mounted at the end of the dipstick. In use the dipstick is dipped into the liquid in which the suspected analyte may be present and allowed to remain for several minutes. The dipstick is then removed and then, either a light is projected through the metalized film or the film is observed with a light behind the film. If a pattern is observed, then the analyte is present in the liquid.

In another embodiment of the present invention, a multiple analyte test is constructed on the same support. As shown in Figure 2, a strip 50 is provided with several micro-contact printed metalized films 70, 75, 80 and 85, each film having a monolayer pattern 60 printed thereon. Each of the micro-contact printed metalized films 70, 75, 80 and 85 have a different receptive material that is different for different analytes. It can be seen that the present invention can be formatted in any array with a variety of micro-contact printed metalized films thereby allowing the user of the biosensor device of the present invention to detect the presence of multiple analytes in a medium using a single test.

In yet another embodiment of the present invention, the biosensor can be attached to an adhesively backed sticker or decal which can then be placed on a hard surface or container wall. The biosensor can be placed on the inside surface of a container such as a food package or a glass vial. The biosensor can then be visualized to determine whether there is microbial contamination.

In one embodiment of the present invention, the receptor layer has the following general formula:

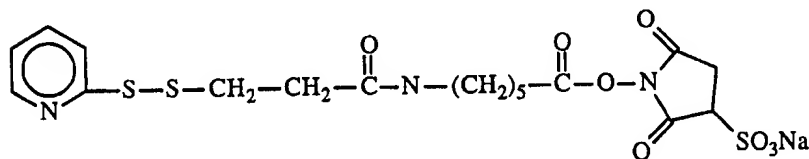
X-R-Y

X is reactive with metal or metal oxide. For example, X may be asymmetrical or symmetrical disulfide (-R'SSR, -RSSR), sulfide (-R'SR, -RSR), diselenide (-R'Se-SeR), selenide (-R'SeR, -RSeR), thiol (-SH), nitrile (-CN), isonitrile, nitro (-NO₂), selenol (-SeH), trivalent phosphorous compounds, isothiocyanate, xanthate, thiocarbamate, phosphine, thioacid or dithioacid, carboxylic acids, hydroxylic acids, and hydroxamic acids.

R is an linker which may optionally be interrupted by hetero atoms and which are preferably non-branched for the sake of optimum dense packing. The linker, helps prevent steric hindrance and/or enhance activity of Y.

Y is the molecule that imparts functionality of the receptor layer. Y can be any molecule that preferentially binds the analyte of interest. Y can be toxins, antibodies, antibody fragments antigens, hormone receptors, parasites, cells, haptens, metabolizes, allergens, nucleic acids, nuclear materials, autoantibodies, blood proteins, cellular debris, enzymes, tissue proteins, enzyme substrates, coenzymes, neuron transmitters, viruses, viral particles, microorganisms, proteins, polysaccharides, chelators, drugs, and any other member of a specific binding pair.

A desired reagent that can be reacted with potential binders such as antibodies or antibody fragments to provide functionality X, include, but are not limited to, sulfo-LC-SPDP (Pierce Chemical Co. Rockford, IL) which has the following formula:



The sulfo-LC-SPDP is reactive towards sulfhydryl and amino groups and is therefore ideally suited for reaction with proteins such as antibodies or other protein receptors, proteoglycans, lipoproteins, glycoproteins, or amino sugars including, but not limited to, glucosamine or galactosamine.

In a typical experimental procedure, schematically shown in Figure 3, a photolithographically produced master is placed in a glass or plastic Petri dish, and a 10:1 ratio (w:w or v:v) mixture of SYLGARD® silicone elastomer 184 and SYLGARD® silicone elastomer 184 curing agent (Dow Corning Corporation) is poured over it. The elastomer is allowed to sit for approximately 30 minutes at room temperature and reduced pressure to degas, then cured for 4 to 16 hours at 60°C, and gently peeled from the master.

"Inking" of the elastomeric stamp is accomplished by soaking the elastomeric stamp in an approximately 0.1 mg/mL to approximately 0.5 mg/mL concentration of the

receptor "ink" for between approximately 10 seconds to 10 minutes, followed by drying the stamp under nitrogen gas. The stamp is allowed to dry until no liquid is visible by eye on the surface of the stamp (typically about 60 seconds), either under ambient conditions, or by exposure to a stream of nitrogen gas. Following inking, the stamp is applied
5 (typically by hand) to a metalized surface. Very light hand pressure is used to aid in complete contact between the stamp and the surface. The stamp should desirably remain on the surface for between approximately 10 seconds to approximately 200 seconds. The actual time the stamp should remain on the surface will vary depending upon the ink used. The stamp is then gently peeled from the surface. A preferred embodiment will follow
10 receptor printing with a passivation step to cover all the surface area of the metal not containing bound receptor. Passivation helps eliminate non-specific binding of analyte.

The elastomeric character of the stamp is important to the success of the process. Polydimethylsiloxane (PDMS), when cured, is sufficiently elastomeric to allow good conformal contact of the stamp and the surface, even for surfaces with significant relief;
15 this contact is essential for efficient contact transfer of the receptor "ink" to the gold film. The elastomeric properties of PDMS are also important when the stamp is removed from the master: if the stamp were rigid (as is the master) it would be difficult to separate the stamp and master after curing without damaging one of the two substrates. PDMS is also sufficiently rigid to retain its shape, even for features with sub-micron dimensions.
20 Patterns have been successfully generated with lines as small as 200 nm in width. The surface of PDMS has a low interfacial free energy ($\gamma = 22.1$ dynes/cm), and the stamp does not adhere strongly to the metalized film. The stamp is durable in that the same stamp can be used up to 100 times over a period of several months without significant degradation in performance. The polymeric nature of PDMS also plays a critical role in the
25 inking procedure, by enabling the stamp to absorb the alkanethiol ink without significant swelling. The stamp can be produced on a printing roll to allow for a continuous printing operation.

A more detailed description of the methods and compositions of the present invention follows. All publications cited herein are incorporated by reference in their
30 entirety.

Any thermoplastic film upon which a metal substrate can be deposited is suitable for the present invention. These include, but are not limited to, polymers such as: polyethylene-terephthalate (MYLAR®), acrylonitrile-butadiene-styrene, acrylonitrile-methyl acrylate copolymer, cellophane, cellulosic polymers such as ethyl cellulose,
35 cellulose acetate, cellulose acetate butyrate, cellulose propionate, cellulose triacetate, cellulose triacetate, polyethylene, polyethylene - vinyl acetate copolymers, ionomers (ethylene polymers) polyethylene-nylon copolymers, polypropylene, methyl pentene

polymers, polyvinyl fluoride, and aromatic polysulfones. Preferably, the plastic film has an optical transparency of greater than 80%. Other suitable thermoplastics and suppliers may be found, for example, in reference works such as the *Modern Plastics Encyclopedia* (McGraw-Hill Publishing Co., New York 1923-1996).

5 In one embodiment of the invention, the thermoplastic film with the metal coating thereon has an optical transparency of between approximately 5% and 95%. A more desired optical transparency for the thermoplastic film used in the present invention is between approximately 20% and 80%. In a desired embodiment of the present invention, the thermoplastic film has at least an approximately 80% optical transparency, and the
10 thickness of the metal coating is such as to maintain an optical transparency greater than about 20%, so that diffraction patterns can be produced by either reflected or transmitted light. This corresponds to a metal coating thickness of about 20 nm. However, in other embodiments of the invention, the gold thickness may be between approximately 1 nm and 1000 nm.

15 The preferred metal for deposition on the film is gold. However, silver, aluminum, chromium, copper, iron, zirconium, platinum and nickel, as well as oxides of these metals, may be used. Chromium oxide can be used to make metalized layers.

 In principle, any surface with corrugations of appropriate size could be used as masters. The process of microcontact printing starts with an appropriate relief structure,
20 from which an elastomeric stamp is cast. This 'master' template may be generated photolithographically, or by other procedures, such as commercially available diffraction gratings. In one embodiment, the stamp may be made from polydimethylsiloxane.

 In one embodiment, the present invention features an optical assay device, having an optically active receptive surface configured and arranged to allow simultaneous assay
25 of a plurality of samples on the surface for one analyte of interest, and an automated liquid handling apparatus (e.g., a pipetting device) configured and arranged to dispense sample and reagent solutions to the surface.

 The present invention has a broad range of applications and, may be utilized in a variety of specific binding pair assay methods. For example, the devices of this invention
30 can be used in immunoassay methods for either antigen or antibody detection. The devices may be adapted for use in direct, indirect, or competitive detection schemes, for determination of enzymatic activity, and for detection of small organic molecules (e.g., drugs of abuse, therapeutic drugs, environmental agents), as well as detection of nucleic acids.

35 The stamp may be applied in air, or under a fluid such as water to prevent excess diffusion of the alkanethiol. For large-scale or continuous printing processes, it is most desirable to print in air, because shorter contact times are desirable for those processes.

In one embodiment of the present invention, the pattern is formed on the metalized thermoplastic polymer with the receptor layer. In another embodiment of the present invention, the relief of the pattern is formed with the receptor layer. After the stamping process, the metalized areas on the plastic may optionally be passivated, for example, with a methyl-terminated monolayer such as hexadecylmercaptan.

This invention is further illustrated by the following examples, which are not to be construed in any way as imposing limitations upon the scope thereof. On the contrary, it is to be clearly understood that resort may be had to various other embodiments, modifications, and equivalents thereof, which, after reading the description herein, may suggest themselves to those skilled in the art without departing from the spirit of the present invention.

EXAMPLE 1

An example derivatization of an antibody to *E. coli* 0157:H7 (Kirkegaard & Perry Labs) follows: To 1 mg of antibody was added 450 μ L phosphate buffered saline (PBS) and 50 μ L of a 10 mM aqueous solution of Sulfo-LC-SPDP (Pierce Catalog #21650). After 60 minutes at room temperature, the solution is desalted in a D-Salt™ Polyacrylamide desalting column (Pierce, Rockford, IL). An acetate buffer, pH 4.5 was used if a subsequent reduction of the disulfide bond was done, while a PBS buffer, pH 7.5, was used if the antibody derivative was to remain as the disulfide. 500 μ L fractions were collected from the column, and the fraction with antibody derivative was determined using a Coomassie® Plus Protein Assay.

EXAMPLE 2

The resulting thiolated antibody from Example 1, either disulfide or thiol, is contact printed on gold-coated MYLAR®. The elastomeric stamp is soaked in a 0.5 mg/mL concentration of the thiolated antibody for 10 minutes, followed by drying the stamp under nitrogen gas, and then contacted with a gold-coated MYLAR® film for 10-120 seconds.

EXAMPLE 3

In this example, a condensation figure is produced. The non-patterned areas after printing as described in Example 2 are reacted with another thiol, such as hexadecanethiol. The condensation figure remained, indicating that the thiolated antibody is chemisorbed and not displaced. An enzyme-linked immunosorbent assay (ELISA) of the printed surface was positive in the patterned areas, verifying the presence of active antibody in the pattern (Figure 4). The ELISA utilized a peroxidase conjugated with an antibody specific for the

E. coli antibody used in Example 1. Tetramethylbenzidine precipitation on the patterned antibody sandwich produced the diffraction pattern. Polystyrene surrogate particles surface modified with antigen also produced patterned adsorption to the receptor layer. The diffraction pattern produced by the tetramethylbenzidine precipitation is shown in Figure 6.

EXAMPLE 4

A surrogate polystyrene particle was produced by carbodiimide coupling with ethyldimethylaminocarbodiimide (EDAC-Sigma Chemical Company, St. Louis, MO) of *E. Coli* 0157:H7 antigen (Kirkegaard & Perry Labs, Cat# 50-95-90) to one micron polystyrene latex spheres by conventional techniques. A 2wt% solution of the diimide was reacted with carboxylate modified PS latex (Bangs: 10^{10} particles/mL) for 4 hours, followed by exposing these activated particles to a 400uG/mL solution of antigen. This surrogate, diluted to 10^8 particles/mL, was exposed to a sensor containing patterned antibody to *E. Coli* 0157:H7, produced as described in Example 2, for 60 minutes. After washing with phosphate buffer, the sample was dried, photographed (Figure 5) and was shown to produce a diffraction pattern as described in Example 3.

EXAMPLE 5

It is well established that one criteria for the presence of a self assembling monolayer (SAM) is increased resistance to chemical etchants and that alkane thiol self assembled monolayers provide resistance of gold to cyanide etching. Cyanide etching was performed to determine if the thiolated protein or oligonucleotide binders of this invention form a protective SAM on gold. Gold coated polyester film (35 nM gold thickness) was exposed to aqueous solutions of either a thiolated antibody to *Candida albicans* (0.5 mG/mL), thiolated protein G (0.5 mG/mL), thiolated oligonucleotide (10 μ M), or underivatized antibody to *Candida albicans* (physisorption only; 0.5 mG/mL); an ethanol solution of hexadecane thiol (HDT; 5.7 mM), known to form a SAM on gold, was used as a positive control. After 16 hours exposure to the thiol containing binders, the coated gold samples were removed, thoroughly rinsed with solvent (water or ethanol), and dried under a nitrogen stream.

Binder coated samples were immersed in an aqueous solution of potassium cyanide (100 mM) containing potassium hydroxide (0.5 M) while bubbling air (oxygen) into the solution. After etching for 11 minutes, the samples were removed, rinsed with water, and visually evaluated for the amount of gold remaining. Table 1 summarizes the observations. The HDT sample was the only sample with most of the gold remaining on

its surface. The thiolated antibody had a very small amount of gold ($\approx 5\%$ coverage), while the other samples had no gold remaining after etching. This demonstrates that unlike HDT, the thiolated binders used to prepare the optical diffraction biosensors do not form a protective SAM.

Table 1 - Summary of Cyanide Etching Experiments

Sample	Observations after etching
Hexadecane thiol (HDT)	70-80%
Thiolated Antibody	$\sim 5\%$ (random specks)
Antibody (physisorbed on surface)	no gold remaining
Thiolated Protein G	no gold remaining
Thiolated Oligonucleotide	no gold remaining

EXAMPLE 6

Samples with patterned antibody to *Candida albicans* were prepared as follows: Gold/polyester (10 nm gold thickness) was pre-treated by immersing it in a 5 mg/mL phosphate-buffered saline solution (pH 7.2) of beta casein (Sigma catalog # C6905) for 10 minutes. The sample was thoroughly rinsed with distilled water, and dried under a strong nitrogen stream. Contact printing was done using a polydimethyl siloxane stamp having an x,y array of 10-micron diameter circles. The stamp was coated with a thiolated antibody to *Candida albicans* (initial polyclonal antibody was Catalog # 20-CR04 from Fitzgerald Industries International, Inc., Concord, MA) by immersing the stamp in a 0.5 mg/mL aqueous solution of the antibody derivative. After 10 minutes, the stamp was removed and thoroughly dried using a strong stream of nitrogen. Contact printing was done on the casein-treated sample, with exposure times of 1 second to 2 minutes being adequate. Two minutes was the preferred contact time. After printing, the sample was again rinsed with distilled water and dried.

The sensor sample was exposed to germ tube-bearing cells of *Candida albicans* by inoculating tape-stripped adult forearm skin with a concentration of 10^6 yeast cells per milliliter, and placing the sensor on top of the yeast containing tape. Transfer of the yeast cells to the sensor was accomplished after only a few seconds of contact (Figure 7). Patterned adhesion of the yeast cells to the sensor was confirmed by microscopic analysis, and resulted in a diffraction image upon irradiation with a laser (Figure 8).

Other surfaces which have been inoculated with germ tube-bearing cells of *Candida albicans* have been an agar plate and a HUGGIES® Baby Wipe. Exposure of these surfaces to the antibody-based sensors has also resulted in patterned attachment of the cells, and diffraction images.

5 Those skilled in the art will now see that certain modifications can be made to the invention herein disclosed with respect to the illustrated embodiments, without departing from the spirit of the instant invention. And while the invention has been described above with respect to the preferred embodiments, it will be understood that the invention is adapted to numerous rearrangements, modifications, and alterations, all such
10 arrangements, modifications, and alterations are intended to be within the scope of the appended claims.

We claim:

1. A biosensor comprising:
a polymer film coated with metal; and
5 a patterned receptor layer printed onto the polymer film wherein the receptor layer has a receptive material thereon that specifically binds an analyte.
- 10 2. The biosensor of Claim 1, wherein the patterned receptor layer is printed in a pattern such that when the biosensor binds the analyte, the biosensor diffracts transmitted or reflected light to form a diffraction pattern.
- 15 3. The biosensor of Claim 2, wherein the diffraction pattern is visible.
- 20 4. The biosensor of Claim 3, wherein the diffraction pattern is visible to the unaided eye.
- 25 5. The biosensor of Claim 2, wherein the diffraction pattern forms a hologram.
- 30 6. The biosensor of Claim 1, wherein the metal is gold, silver, chromium, nickel, platinum, aluminum, iron, copper, chromium oxide or zirconium.
7. The biosensor of Claim 6, wherein the metal is gold.
8. The biosensor of Claim 6, wherein the metal coating is between approximately 1 nanometer and 1000 nanometers in thickness.

5 9. The biosensor of Claim 1, wherein the polymer film is polyethylene-terephthalate, acrylonitrile-butadiene-styrene, acrylonitrile-methyl acrylate copolymer, cellophane, cellulosic polymers such as ethyl cellulose, cellulose acetate, cellulose acetate butyrate, cellulose propionate, cellulose triacetate, cellulose triacetate, polyethylene, polyethylene - vinyl acetate copolymers, ionomers (ethylene polymers) polyethylene-nylon copolymers, polypropylene, methyl pentene polymers, polyvinyl fluoride, and aromatic polysulfones.

10 10. The biosensor of Claim 9, wherein the polymer film is polyethylene-terephthalate.

15 11. The biosensor of Claim 1, wherein the thermoplastic film is optically transparent.

20 12. The biosensor of Claim 1, wherein the thermoplastic film has an optical transparency between 5% and 95%.

25 13. The biosensor of Claim 1, wherein the thermoplastic film has an optical transparency between approximately 20% and 80%.

30 14. The biosensor of Claim 1, wherein the patterned receptor layer is formed from compounds with the following general formula:



35

wherein:

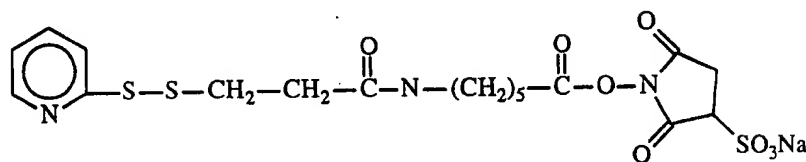
X is reactive with the metal or metal oxide on the polymer film;

R is an optional linker; and

Y is a compound with any property of interest.

15. The biosensor of Claim 14, wherein R is between 0 and 12 carbon atoms in length.

16. The biosensor of Claim 14, wherein X is generated from a compound comprising the following formula:



17. The biosensor of Claim 1, wherein the analyte is bacteria, yeast, fungus, virus, rheumatoid factor, IgG, IgM, IgA and IgE antibodies, carcinoembryonic antigen, streptococcus Group A antigen, viral antigens, antigens associated with autoimmune disease, allergens, tumor antigens, streptococcus Group B antigen, HIV I or HIV II antigen, antibodies viruses, antigens specific to RSV., an antibody, antigen, enzyme, hormone, polysaccharide, protein, lipid, carbohydrate, drug or nucleic acid, *Neisseria meningitidis* groups A, B, C, Y and W sub 135, *Streptococcus pneumoniae*, *E. coli K1*, *Haemophilus influenza* type B, an antigen derived from microorganisms, a hapten, a drug of abuse, a therapeutic drug, an environmental agents, or antigens specific to Hepatitis.

18. The biosensor of Claim 17, wherein the analyte is bacteria, yeast, fungus or virus.

19. The biosensor of Claim 1, wherein the receptor material is antigens, antibodies, nucleotides, chelators, enzymes, bacteria, yeasts, fungi, viruses, bacterial pili, bacterial flagellar materials, nucleic acids, polysaccharides, lipids, proteins, carbohydrates, metals, hormones and receptors for said materials.

20. The biosensor of Claim 19, wherein the fungus is *Candida species*.

21

21. The biosensor of Claim 19, wherein the bacteria is *Salmonella species*.

5 22. The biosensor of Claim 1, wherein the biosensor is attached to the inside wall of a container.

23. The biosensor of Claim 22, wherein the container is a vial.

10 24. The biosensor of Claim 21, wherein the container is a food container.

15 25. The biosensor of Claim 1, wherein the biosensor is attached to the inside wall of a garment.

26. The biosensor of Claim 24, wherein the garment is a diaper.

20 27. The biosensor of Claim 1, wherein the analyte is attached to a particle.

25 28. The biosensor of Claim 27, wherein the particle is comprised of glass, cellulose, latex, polystyrene, polycarbonate, protein, or microbial cells.

30 29. The biosensor of Claim 27, wherein the particle is between approximately 0.2 nm and 50 nm.

30. The biosensor of Claim 29, wherein the particle is between approximately 0.4 μm to 1 μm .

31. The biosensor of Claim 29, wherein the particle size is determined by the following formula:

$$t_{\text{opt}} = \lambda/2(n_2 - n_1)$$

wherein t_{opt} = optimum height of the particle
 λ = wavelength of incoming light
 n_2 = refractive index of particle
 n_1 = refractive index of surrounding medium.

32. A method of detecting an analyte comprising
contacting the analyte with a biosensor, the biosensor comprising:
a polymer film coated with metal; and
a patterned receptor layer printed onto the polymer film wherein the
receptor layer has a receptive material thereon that specifically
binds the analyte;
transmitting light through the biosensor with the analyte bound to the
patterned receptor layer thereby forming a diffraction pattern.

33. The method of Claim 32, wherein the light is reflected from the biosensor.

34. The method of Claim 32, wherein the diffraction pattern is visible.

35. The method of Claim 34, wherein the diffraction pattern is visible to the unaided eye.

36. The method of Claim 32, wherein the diffraction pattern forms a hologram.

37. The method of Claim 32, wherein the metal is selected from the group consisting of gold, silver, chromium, nickel, platinum, aluminum, iron, copper, chromium oxide or zirconium.

38. The method of Claim 37, wherein the metal is gold.

5

39. The method of Claim 37, wherein the metal coating is between approximately 1 nanometer and 1000 nanometers in thickness.

10

40. The method of Claim 32, wherein the polymer film is polyethylene-terephthalate, acrylonitrile-butadiene-styrene, acrylonitrile-methyl acrylate copolymer, cellophane, cellulosic polymers such as ethyl cellulose, cellulose acetate, cellulose acetate butyrate, cellulose propionate, cellulose triacetate, cellulose triacetate, polyethylene, polyethylene - vinyl acetate copolymers, ionomers (ethylene polymers) polyethylene-nylon

15

copolymers, polypropylene, methyl pentene polymers, polyvinyl fluoride, and aromatic polysulfones.

20

41. The method of Claim 40, wherein the polymer film is polyethylene-terephthalate.

25

42. The method of Claim 32, wherein the thermoplastic film is optically transparent.

30

43. The method of Claim 32, wherein the thermoplastic film has an optical transparency between 5% and 95%.

44. The method of Claim 32, wherein the thermoplastic film has an optical transparency between approximately 20% and 80%.

24

45. The film of Claim 32, wherein the patterned receptor layer is formed from compounds with the following general formula:



wherein:

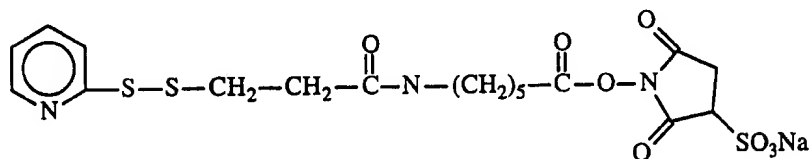
X is reactive with the metal or metal oxide on the polymer film;

R is an optional linker; and

Y is a compound with any property of interest.

46. The method of Claim 45, wherein R is between 0 and 12 carbon atoms in length.

47. The method of Claim 45, wherein X is generated from a compound comprising the following formula:



48. The method of Claim 32, wherein the analyte is bacteria, yeast, fungus, virus, rheumatoid factor, IgG, IgM, IgA and IgE antibodies, carcinoembryonic antigen, streptococcus Group A antigen, viral antigens, antigens associated with autoimmune disease, allergens, tumor antigens, streptococcus Group B antigen, HIV I or HIV II antigen, antibodies viruses, antigens specific to RSV,, an antibody, antigen, enzyme, hormone, polysaccharide, protein, lipid, carbohydrate, drug or nucleic acid, *Neisseria meningitides* groups A, B, C, Y and W sub 135, *Streptococcus pneumoniae*, *E. coli* K1, *Haemophilus influenza* type B, an antigen derived from microorganisms, a hapten, a drug of abuse, a therapeutic drug, an environmental agents, or antigens specific to Hepatitis.

49. The method of Claim 48, wherein the analyte is bacteria, yeast, fungus or virus.

50. The method of Claim 49, wherein the fungus is *Candida species*.

5 51. The method of Claim 49, wherein the bacteria is *Salmonella species*.

10 52. The method of Claim 32, wherein the receptor material is antigens, antibodies, nucleotides, chelators, enzymes, bacteria, yeasts, fungi, viruses, bacterial pili, bacterial flagellar materials, nucleic acids, polysaccharides, lipids, proteins, carbohydrates, metals, hormones and receptors for said materials.

15 53. The method of Claim 32, wherein the biosensor is attached to the inside wall of a container.

54. The method of Claim 53, wherein the container is a vial.

20 55. The method of Claim 54, wherein the container is a food container.

25 56. The method of Claim 32, wherein the biosensor is attached to the inside wall of a garment.

57. The method of Claim 56, wherein the garment is a diaper.

30 58. The method of Claim 32, wherein the analyte is attached to a particle.

35 59. The method of Claim 58, wherein the particle is comprised of glass, cellulose, latex, polystyrene, polycarbonate, protein, or microbial cells.

60. The method of Claim 58, wherein the particle is between approximately 0.2 nm and 50 nm.

61. The method of Claim 60, wherein the particle is between approximately 0.4 μm to 1 μm .

62. The method of Claim 58, wherein the particle size is determined by the following formula:

$$t_{\text{opt}} = \lambda/2(n_2 - n_1)$$

wherein t_{opt} = optimum height of the particle

λ = wavelength of incoming light

n_2 = refractive index of particle

n_1 = refractive index of surrounding medium.

63. A method of making a biosensor comprising applying a receptor layer to a polymer film coated with metal in a pattern such that after an analyte binds to the patterned receptor layer, the patterned receptor layer thereby forms a diffraction image.

- 5 64. A method of detecting an analyte comprising
- a. contacting the analyte with a biosensor, the biosensor comprising:
a polymer film coated with metal; and
a patterned receptor layer printed onto the polymer film wherein
the receptor layer has a first receptive material thereon that
specifically binds the analyte to form an analyte/first
receptive material conjugate;
- 10 b. contacting the biosensor with the analyte/first receptive material
conjugate with a second receptor material that specifically binds the
analyte/first receptive material conjugate, wherein the second receptor
material is conjugated to a precipitate forming material;
- c. contacting the biosensor from step b with a reagent that will cause a
precipitate to form;
- 15 d. transmitting light through the biosensor with the analyte bound to the
patterned receptor layer thereby forming a diffraction pattern.
- 20 65. The method of Claim 64, wherein the precipitate forming material is a
peroxidase enzyme or colloidal gold.
- 25 66. The method of Claim 64, wherein the reagent is tetramethylbenzidine or a
silver halide.
- 30 67. The method of Claim 64, wherein the second receptor material is specific
for the first receptor material.
68. The method of Claim 64, wherein the first receptor material is an antibody
conjugated to an enzyme.
- 35 69. The method of Claim 64, wherein the second receptor material is an
antibody.

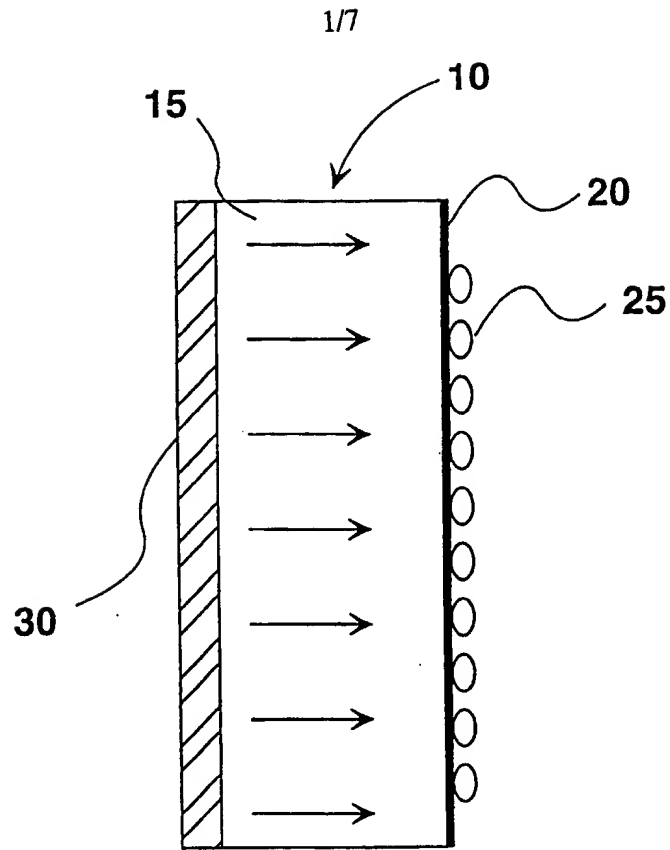


Figure 1

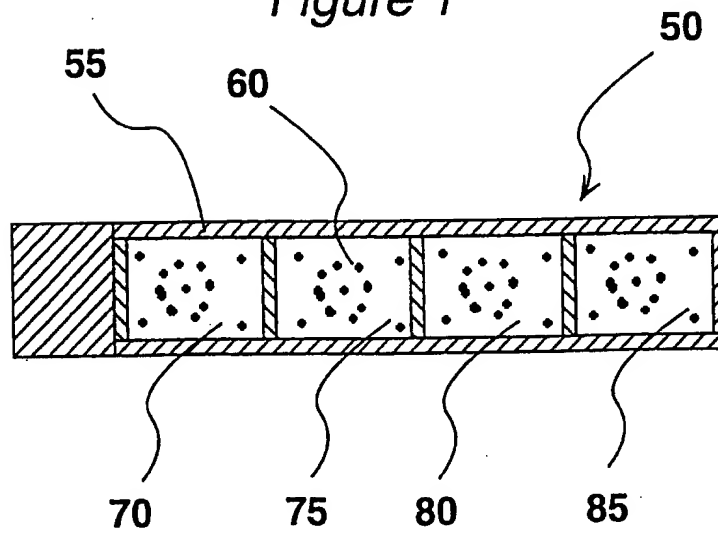
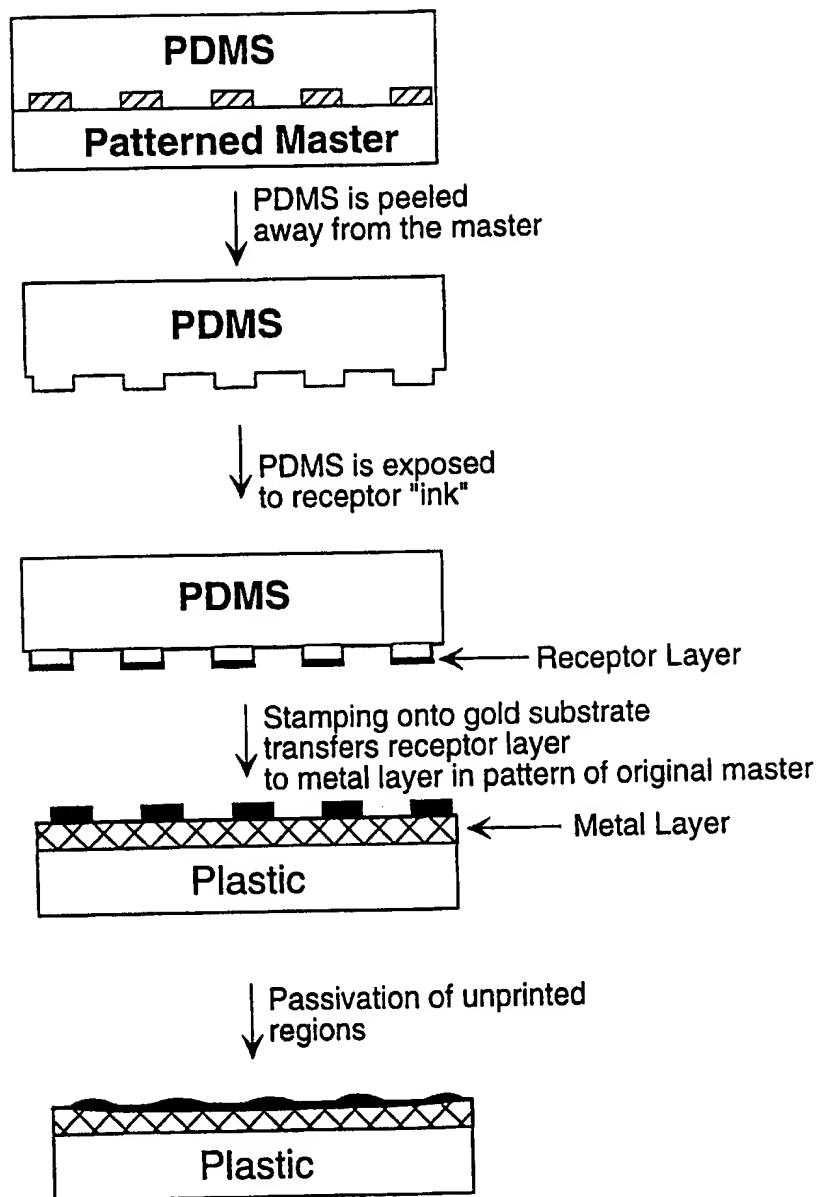


Figure 2

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*Figure 3*

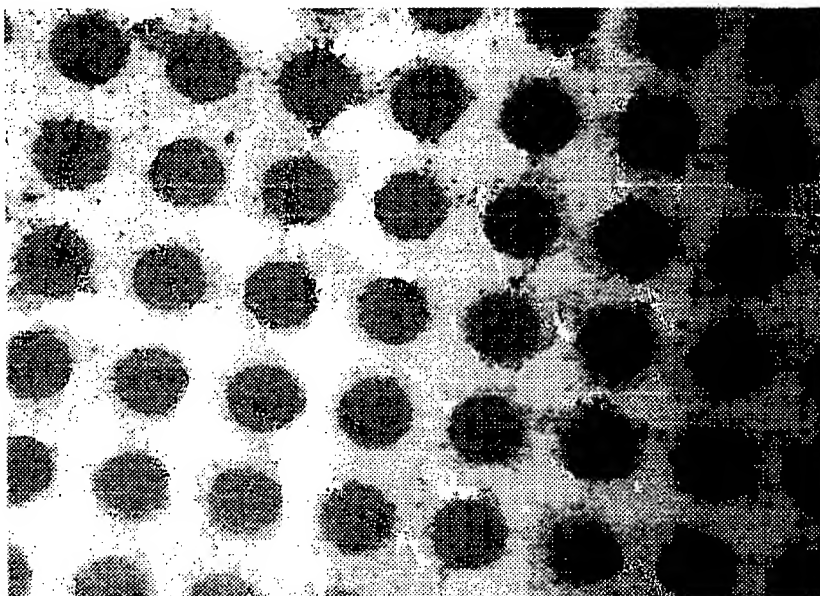


Figure 4

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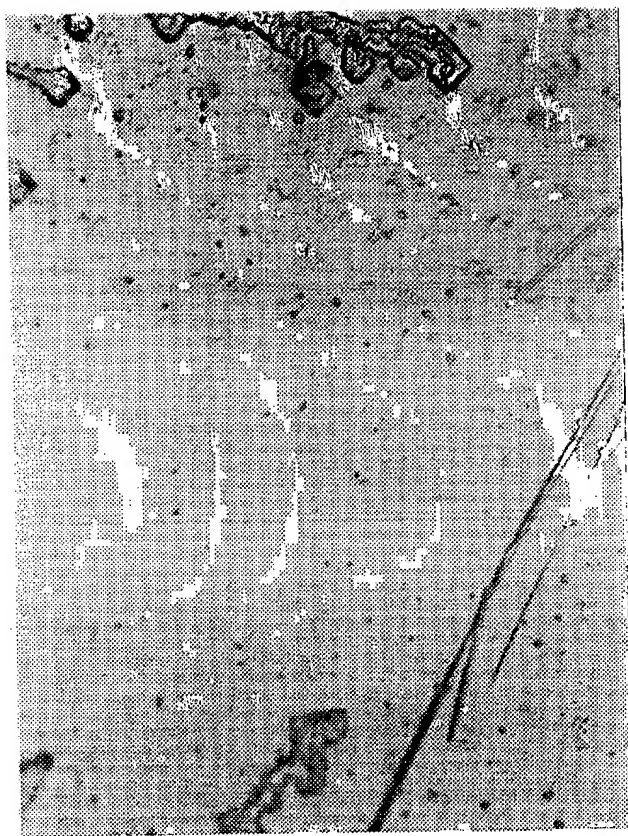


Figure 5

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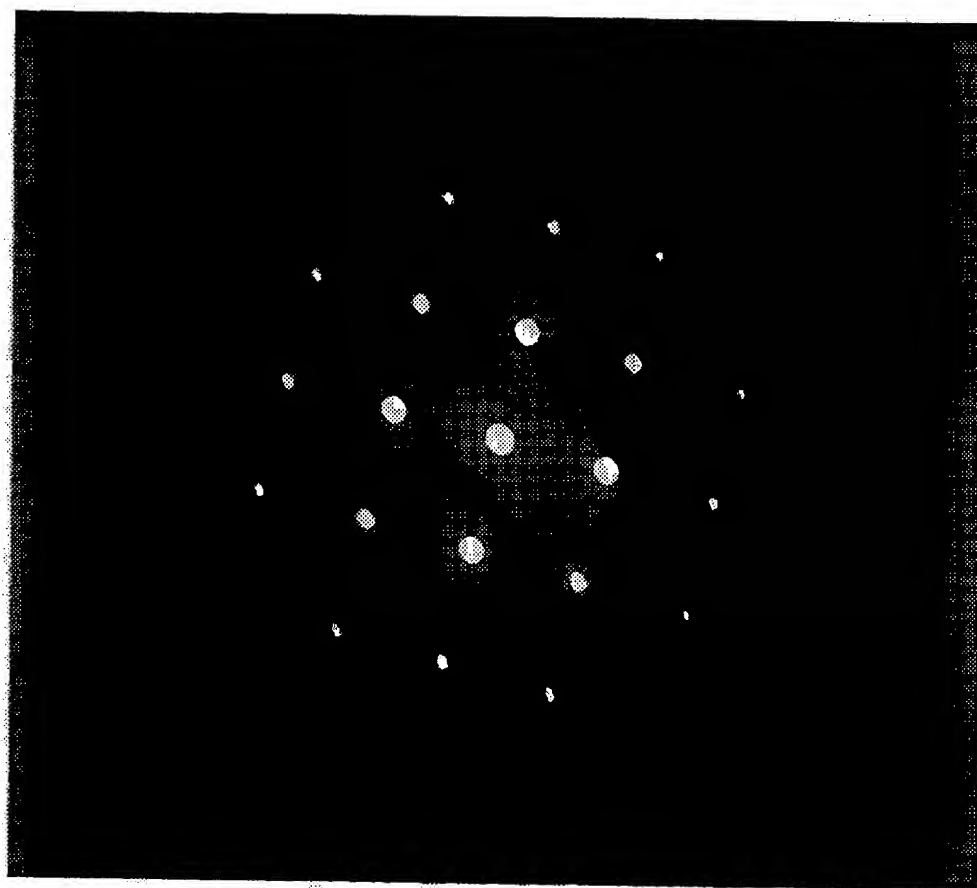


Figure 6

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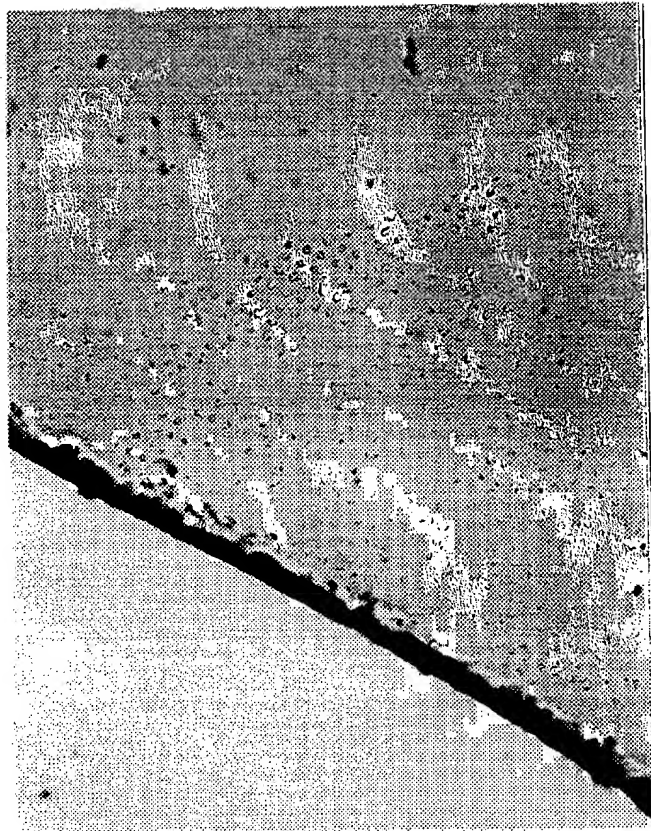


Figure 7

7/7

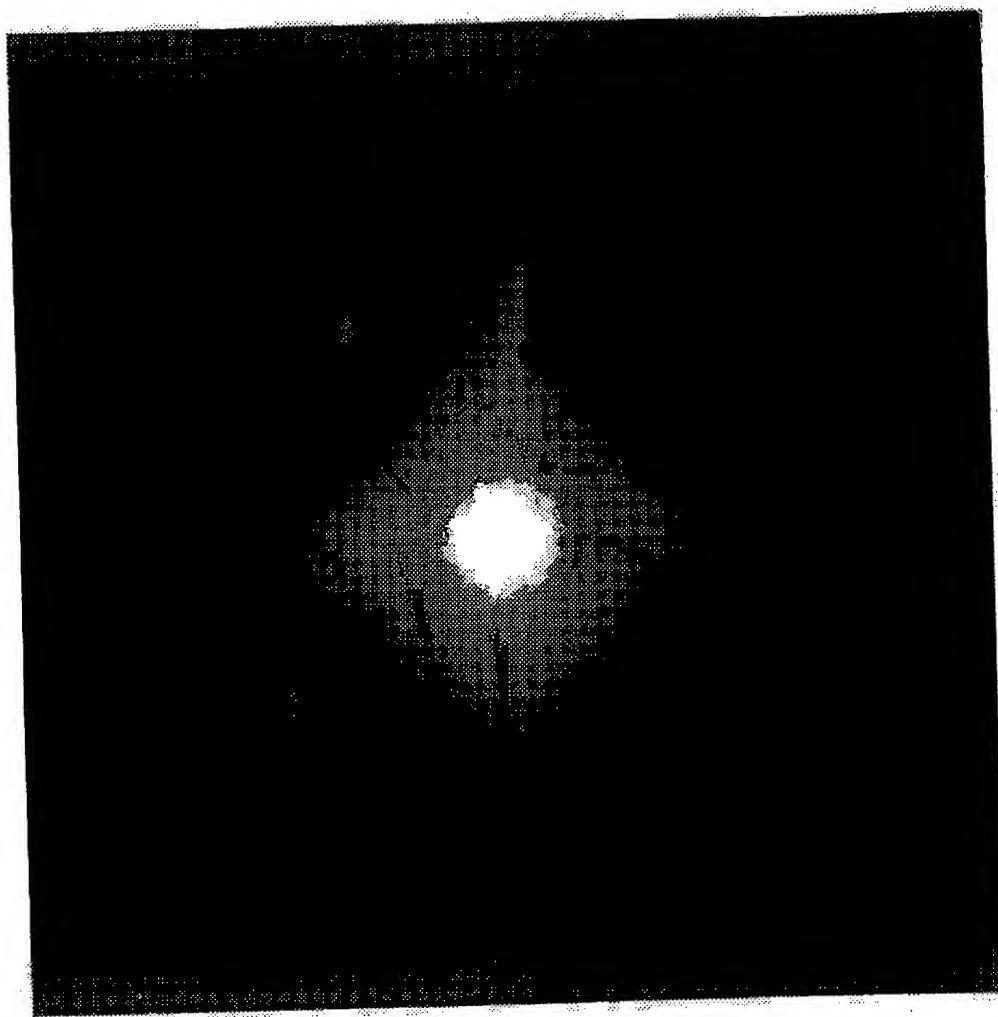


Figure 8

INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 98/26759

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 G01N21/47 B41M3/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 G01N B41M G03F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5 512 131 A (WHITESIDES GEORGE M ET AL) 30 April 1996 see column 15, line 52 - column 16, line 30; figure 1 ---	1-3, 32-35,63
A	MRKSICH M ET AL: "PATTERNING SELF-ASSEMBLED MONOLAYERS USING MICROCONTACT PRINTING: A NEW TECHNOLOGY FOR BIOSENSORS" TIBTECH, vol. 13, June 1995, pages 228-235, XP002060826 see page 230, right-hand column, paragraph 2; figure 2 --- -/--	1-3, 32-35,63

☒ Further documents are listed in the continuation of box C.

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Date of the actual completion of the international search

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Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 96 29629 A (HARVARD COLLEGE) 26 September 1996 see page 2, line 16 - line 25 see abstract; figure 1 see page 16, line 24 - page 17, line 12 ----	1-3, 32-35,63
A	US 5 196 350 A (BACKMAN KEITH C ET AL) 23 March 1993 see abstract ----	2,32,63
P,X	WO 98 10334 A (KIMBERLY CLARK CO) 12 March 1998 see the whole document ----	1,2, 6-15, 32-35, 37-45,63
P,X	WO 98 27417 A (KIMBERLY CLARK CO) 25 June 1998 see the whole document -----	1-15, 17-30, 32-46, 48-61,63

INTERNATIONAL SEARCH REPORT

information on patent family members

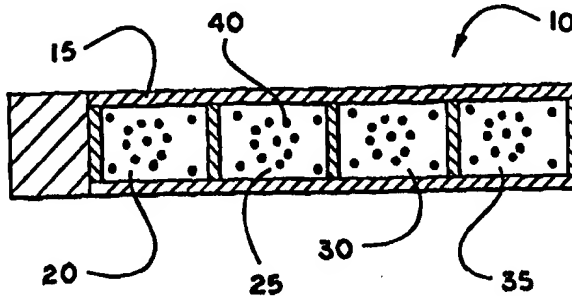
International Application No

PCT/US 98/26759

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WO 9629629	A	26-09-1996	EP 0812434 A	17-12-1997
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			AU 670252 B	11-07-1996
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<p>(21) International Application Number: PCT/US99/27671</p> <p>(22) International Filing Date: 22 November 1999 (22.11.99)</p> <p>(30) Priority Data: 09/210,016 11 December 1998 (11.12.98) US</p> <p>(71) Applicant: KIMBERLY-CLARK WORLDWIDE, INC. [US/US]; 401 North Lake Street, Neenah, WI 54956 (US).</p> <p>(72) Inventors: EVERHART, Dennis, S.; 230 Hereford Road, Alpharetta, GA 30004 (US). KAYLOR, Rosann, M.; 7480 Williamsburg Drive, Cumming, GA 30041 (US). MCGRATH, Kevin; 335 Hermitage Trail, Alpharetta, GA 30004 (US).</p> <p>(74) Agents: GREEN, Theodore, M. et al.; Jones & Askew, LLP, 2400 Monarch Tower, 3424 Peachtree Road, N.E., Atlanta, GA 30326 (US).</p>		<p>(81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).</p> <p>Published <i>Without international search report and to be republished upon receipt of that report.</i></p>
<p>(54) Title: PATTERNED BINDING OF FUNCTIONALIZED MICROSPHERES FOR OPTICAL DIFFRACTION-BASED BIOSENSORS</p> <p>(57) Abstract</p> <p>The present invention provides an inexpensive and sensitive system and method for detecting analytes present in a medium. The system comprises a diffraction enhancing element, such as functionalized microspheres, which are modified such that they are capable of binding with a target analyte. Additionally, the system comprises a polymer film, which may include a metal coating, upon which is printed a specific, pre-determined pattern of analyte-specific receptors. Upon attachment of a target analyte to select areas of the polymer film, either directly or with the diffraction enhancing element, diffraction of transmitted and/or reflected light occurs via the physical dimensions and defined, precise placement of the analyte. A diffraction image is produced which can be easily seen with the eye or, optionally, with a sensing device.</p> 		

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**PATTERNED BINDING OF FUNCTIONALIZED
MICROSPHERES FOR OPTICAL DIFFRACTION-BASED
BIOSENSORS**

10 TECHNICAL FIELD

The present invention is generally in the field of detecting analytes in a medium and, more particularly, the present invention relates to the use of functionalized microspheres for enhancing optical diffraction with single use, disposable sensors to indicate the presence of the analyte in a medium.

15

BACKGROUND OF THE INVENTION

There are many systems and devices available for detecting a wide variety of analytes in various media. Most of these systems and devices are relatively expensive and require a trained technician to perform the test. There are many cases where it would be advantageous to be able to rapidly and inexpensively determine if an analyte were present. What is needed is a biosensor system that is easy and inexpensive to manufacture and is capable of reliable and sensitive detection of analytes, including smaller analytes.

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Sandstrom et al., 24 *Applied Optics* 472, 1985, describe use of an optical substrate of silicon with a layer of silicon monoxide and a layer of silicon formed as dielectric films. They indicate that a change in film thickness changes the properties of the optical substrate to produce different colors related to the thickness of the film. The thickness of the film is related to the color observed and a film provided on top of an optical substrate may produce a visible color change. The authors indicate that a mathematical model can be used to quantitate the color change, and that "[c]alculations performed using the computer model show that very little can be gained in optical performance from using a multilayer structure... but a biolayer on the surface

30

35

changes the reflection of such structures very little since the optical properties are determined mainly by the interfaces inside the multilayer structure. The most sensitive system for detection of biolayers is a single layer coating, while in most other applications performance can be by additional dielectric layers.”

Sandstrom et al., go on to indicate that slides formed from metal oxides on metal have certain drawbacks, and that the presence of metal ions can also be harmful in many biochemical applications. They indicate that the ideal top dielectric film is a 2-3 nm thickness of silicon dioxide which is formed spontaneously when silicon monoxide layer is deposited in ambient atmosphere, and that a 70-95 nm layer silicon dioxide on a 40-60 nm layer of silicon monoxide can be used on a glass or plastic substrate. They also describe formation of a wedge of silicon monoxide by selective etching of the silicon monoxide, treatment of the silicon dioxide surface with dichlorodimethylsilane, and application of a biolayer of antigen and antibody. From this wedge construction they were able to determine film thickness with an ellipsometer, and note that the “maximum contrast was found in the region about 65 nm where the interference color changed from purple to blue.” They indicate that the sensitivity of such a system is high enough for the detection of protein antigen by immobilized antibodies. They conclude “the designs given are sensitive enough for a wide range of applications. The materials, i.e., glass, silicon, and silicon oxides, are chemically inert and do not affect the biochemical reaction studied. Using the computations above it is possible to design slides that are optimized for different applications. The slides can be manufactured and their quality ensured by industrial methods, and two designs are now commercially available.

U.S. Patent 5,512,131 issued to Kumar et al. describes a device that includes a polymer substrate having a metal coating. An analyte-specific receptor layer is stamped on the coated substrate. The device is used in a process for stamping or as a switch. A diffraction pattern is generated when an analyte binds

to the device. A visualization device, such as a spectrometer, is then used to determine the presence of the diffraction pattern.

5 However, the device described by Kumar et al. has several disadvantages. One disadvantage is that an extra visualization device is needed to view any diffraction pattern. By requiring a visualization device, the Kumar et al. device does not allow a large number of samples to be tested since it is not possible to determine the presence of an analyte by using the unaided eye. Additionally, this device is not able to detect smaller analytes as
10 these analytes do not produce a noticeable diffraction pattern.

U.S. Patent No. 5,482,830 to Bogart, et al., describes a device that includes a substrate which has an optically active surface exhibiting a first color in response to light impinging thereon. This first color is defined as a spectral distribution of
15 the emanating light. The substrate also exhibits a second color which is different from the first color (by having a combination of wavelengths of light which differ from that combination present in the first color, or having a different spectral distribution, or by having an intensity of one or more of those
20 wavelengths different from those present in the first color). The second color is exhibited in response to the same light when the analyte is present on the surface. The change from one color to another can be measured either by use of an instrument, or by eye. Such sensitive detection is an advance over the devices
25 described by Sandstrom and Nygren, supra, and allow use of the devices in commercially viable and competitive manner.

However, the method and device described in the Bogart, et al. patent has several disadvantages. One disadvantage is the high cost of the device. Another problem with the device is the
30 difficulty in controlling the various layers that are placed on the wafer so that one obtains a reliable reading.

Additionally, biosensors having a self-assembling monolayer have been used to detect analytes and are set forth in
35 U.S. Patent Application Nos. 08/768,449 and 08/991,844, both of which are incorporated herein by reference in their entirety. However, these biosensors currently do not have the requisite sensitivity required to detect smaller analytes since these smaller

analytes do not produce a sufficient diffraction pattern to be visible.

5 Some commercial lateral flow technologies have been used which employ latex bead technology. These technologies are currently employed in most of the commercially-available home diagnostic kits (e.g. pregnancy and ovulation kits). These kits use colored beads which accumulate in a defined "capture zone" until the amount of beads becomes visible to the unaided eye. However, these systems lack the requisite sensitivity to test
10 for many analytes, since a much larger number of latex beads must bind in the capture zone to be visible to the naked eye than that required to cause diffraction in the same size zone. Generally, the number of beads needed is about 2 to 3 orders of magnitude higher than the sensors of the present invention.

15 What is needed is a biosensor system that is easy and inexpensive to manufacture and is capable of reliable and sensitive detection of analytes, including smaller analytes.

SUMMARY OF THE INVENTION

20 The present invention provides an inexpensive and sensitive system and method for detecting analytes present in a medium. The system comprises a biosensing device having a polymer film upon which is printed a specific, predetermined pattern of analyte-specific receptors. The polymer film may be
25 coated with a metal layer. Additionally, the system utilizes "diffraction enhancing elements" which are capable of binding to the target analyte and to the biosensor and are capable of producing a substantial change in the height and/or refractive index, thereby increasing the diffraction efficiency of the
30 biosensor and permitting the detection of smaller analytes. In use, a target analyte attaches either to the diffraction enhancing element, which then attaches to the biosensor, or directly to select areas of the polymer film upon which the receptor is printed. Then diffraction of transmitted and/or reflected light
35 occurs via the physical dimensions and defined, precise placement of the analyte. A diffraction image is produced which

can be easily seen with the eye or, optionally, with a sensing device.

5 The system of the present invention is much more sensitive than current inexpensive systems. The system of the present invention is able to detect low to high molecular weight analytes, microorganisms, and DNA or RNA species in fluid samples. More specifically, the system is able to detect hormones, steroids, antibodies, drug metabolites, and even nucleic acids, among others. This is a significant expansion of
10 the optical diffraction-based sensing technology set forth in U.S. Patent Application Nos. 08/768,449 and 08/991,844.

The present invention utilizes diffraction enhancing elements, such as latex microspheres, which aid in the detection of smaller analytes. Normally, after an analyte binds to an
15 analyte-specific receptor on a biosensor, the analyte will diffract or reflect transmitted light to produce a diffraction pattern. If the analyte is larger, the diffraction pattern is able to be seen with the unaided eye. However, some analytes are too small such that the diffraction pattern produced is not able to be seen. By using
20 diffraction enhancing elements, the biosensor having the analyte-specific receptor material may be used to detect these smaller analytes. The diffraction enhancing elements used are capable of binding to the analyte, and then the element with bound analyte binds to the biosensor. Then, as the light is transmitted through
25 or reflected from the biosensor, the element enhances the diffraction pattern generated by the analyte such that the resulting diffraction pattern may be seen by the unaided eye.

The present invention also utilizes methods of contact printing of patterned, analyte-specific receptors. The analyte-specific receptors have receptive materials bound thereto. The
30 receptive materials are specific for a particular analyte or class of analyte, depending upon the receptor used. Methods of contact printing which would be useful in generating the sensing devices used in the present system are disclosed fully in U.S. Patent
35 Application Nos. 08/707,456 and 08/769,594, both of which are incorporated herein by reference in their entirety. However, since these methods relate to self-assembling monolayers, the

methods need to be altered slightly, as discussed below, to print the analyte-specific receptor material as this material is not self-assembling.

5 Patterned analyte-specific receptor layers allow for the controlled placement of analytes and/or diffraction enhancing elements thereon via the patterns of analyte-specific receptors. The biosensing devices of the present invention produced thereby are used by first exposing the biosensing device to a medium that contains the analyte of choice mixed with the
10 diffraction enhancing element. Then, after an appropriate incubation period, a light, such as a laser or other point light source, is transmitted through or reflected from the film. If the analyte is present in the medium and is bound, either directly or in conjunction with the diffraction enhancing element, to the
15 receptors on the patterned analyte-specific receptor layer, the light is diffracted in such a way as to produce a visible image. In other words, the analyte-specific receptor layers with the analyte and/or diffraction enhancing element bound thereto can produce optical diffraction patterns which differ depending on
20 the reaction of the receptors on the analyte-specific receptor layer with the analyte of interest. The light can be in the visible spectrum, and be either reflected from the film, or transmitted through it, and the analyte can be any compound or particle reacting with the analyte-specific receptor layer. The light can be
25 a white light or monochromatic electromagnetic radiation in the visible region. While visible light is the desired light source, the present invention may also be used with non-visible point light sources, such as near-infrared light, coupled with a detector. The thickness of the film and the size of the microparticle may be
30 adjusted to compensate for the non-visible light source. Additionally, the present invention also provides a flexible support for an analyte-specific receptor layer either directly on the substrate or on gold or other suitable metal or metal alloy.

35 The present invention provides an analyte-specific receptor layer on gold or other material which is suitable for mass production. The biosensors used in the present invention can be produced as a single test for detecting an analyte or it can be

formatted as a multiple test device. The biosensors of the present invention can be used to detect (1) antigens or antibodies associated with medical conditions, (2) contamination in garments, such as diapers, and (3) contamination by microorganisms.

In another embodiment of the present invention, nutrients for a specific class of microorganisms can be incorporated into the analyte-specific receptor layer. In this way, very low concentrations of microorganisms can be detected by first contacting the biosensor of the present invention with the nutrients incorporated therein and then incubating, if necessary, the biosensor under conditions appropriate for the growth of the bound microorganism. The microorganism is allowed to grow until there are enough organisms to form a diffraction pattern.

The present invention can also be used on contact lenses, eyeglasses, window panes, pharmaceutical vials, solvent containers, water bottles, adhesive bandages, and the like to detect contamination.

These and other features and advantages of the present invention will become apparent after a review of the following detailed description of the disclosed embodiments.

BRIEF DESCRIPTION OF THE FIGURES

Figure 1 shows a biosensor capable of simultaneously measuring several different analytes in a medium.

Figure 2 is a schematic of contact printing of analyte-specific receptor layers.

Figure 3 is an atomic force microscopy image of evaporated gold on MYLAR®, purchased from Courtaulds Performance Films (Canoga Park, CA). The average roughness of the gold layer is 3-4 nanometers, with maximum roughness of 9 nanometers.

Figure 4 is an SEM photomicrograph showing patterned attachment of diffraction enhancing elements in the presence of an analyte.

DETAILED DESCRIPTION

5 The present invention features improved biosensing devices, and methods for using such biosensing devices, for detecting and quantifying the presence or amount of an analyte of interest within a medium. The present invention is much more sensitive and can be used to detect smaller analytes which, until now, were not able to be detected without the use of expensive instruments. The analytes that can be detected by the present invention include, but are not limited to, hormones, 10 proteins such as antibodies, steroids, drug metabolites, nucleic acids, microorganisms such as bacteria, yeasts, fungi and viruses. In contrast to prior devices, those of the present invention allow detection of extremely small quantities and sizes of analytes in a medium in a rapid assay lasting only a few minutes. In addition, 15 no signaling or associated electronic components are required in the present invention.

The present invention comprises micro-contact printing of analyte-specific receptors onto polymer film, which may have a metal coating thereon. The invention allows for the development 20 of single use, disposable biosensors based on light diffraction to indicate the presence of the analyte. Additionally, the present invention includes diffraction enhancing elements which increase the diffraction efficiency of the biosensor, thereby making it possible to detect any number of different analytes. Upon 25 attachment of a target analyte to select areas of the polymer film which contain the receptor, either directly or in combination with a diffraction enhancing element, diffraction of transmitted and/or reflected light occurs via the physical dimensions and defined, precise placement of the analyte. For example, yeast, fungi or 30 bacterium are large enough to act as diffraction elements for visible light when placed in organized patterns on a surface. However, smaller analytes, such as viruses, proteins, molecules, hormones, steroids, drug metabolites and nucleic acids, are only capable of acting as suitable diffraction elements when they are 35 also bound to a diffraction enhancing element. In addition to producing a simple diffraction image, patterns of analytes can be such as to allow for the development of a holographic sensing

image and/or a change in visible color. Thus, the appearance of a hologram or a change in an existing hologram will indicate a positive response. The pattern made by the diffraction of the transmitted light can be any shape including, but not limited to, the transformation of a pattern from one pattern to another upon binding of the analyte to the receptive material. In particularly preferred embodiments, the diffraction pattern is discernible in less than one hour after contact of the analyte with the biosensing device of the present invention.

The diffraction grating which produces the diffraction of light upon interaction with the analyte and/or element should have a minimum periodicity of the wavelength of incident light. Very small analytes can be detected indirectly by using diffraction enhancing element particles that are specific for the small analyte. One embodiment in which the small analyte can be detected comprises coating the element particle, such as a latex bead, with a receptor material that specifically binds to the analyte of interest.

A variety of methods may be used to attach the receptor material onto the diffraction enhancing particle. These methods include, but are not limited to, simple physisorption to a hydrophobic particle (e.g., binding a protein onto polystyrene particles); binding using a protein A or protein G linker; binding using a streptavidin or avidin-biotin linker; or binding using covalent attachment. A preferred embodiment of the present invention is to use carbodiimide coupling of a proteinaceous receptor to carboxylated particles. Other methods of coupling well-known to those of ordinary skill in the art may be used as well.

Diffraction enhancing element particles that can be used in the present invention include, but are not limited to, glass, cellulose, synthetic polymers or plastics, latex, polystyrene, polycarbonate, bacterial or fungal cells and the like. The particles are preferably spherical in shape, but the structural and spatial configuration of the particle is not critical to the present invention. For instance, the particles could be slivers, ellipsoids, cubes, and the like. A desirable particle size ranges from a

diameter of approximately 0.1 μm to 100.0 μm , desirably between approximately 0.3 μm to 1 μm . The composition of the element particle is not critical to the present invention. Preferably, the difference in refractive index between the medium and the enhancing element is between 0.1 and 1.0. More preferably, the difference in refractive index between the medium and the enhancing element is between 0.2 and 0.7

The analyte-specific receptor layer on the polymer film contains a receptive material, such as an antibody, that will specifically bind to an epitope on the analyte that is different from the epitope used in the binding to the particle. Thus, for detecting a small analyte, such as viral particles, the medium is first exposed to the diffraction enhancing element particles, such as latex particles, to which the viral particles bind. Then, the diffraction enhancing element particles are optionally washed and exposed to the polymer film with the analyte-specific receptor layers containing the virus specific antibodies. The antibodies then bind to the viral particles on the element particle thereby immobilizing the element particles in the same pattern as the receptors on the film. Because the bound element particles will cause diffraction of the visible light, a diffraction pattern is formed, indicating the presence of the viral particle in the liquid. Additionally, the polymer film may include a metal coating thereon. The analyte-specific receptor layer would then be located on the metalized surface of the film.

Alternatively, the analyte may be detected by first exposing the substrate to the medium containing the analyte and causing the analyte to bind to the analyte-specific receptor layer material. Next, a solution containing the diffraction enhancing element particles is contacted with the substrate having the analyte bound thereto. The particles then bind to the analyte. Because the bound element particles will cause diffraction of the visible light, a diffraction pattern is formed, indicating the presence of the analyte in the liquid.

Finally, in a preferred embodiment, the biosensor, the diffraction enhancing element particles and the medium containing the analyte may be admixed simultaneously. This will

5 result in a combination of the binding procedures discussed above. Some of the analytes will first bind with a diffraction enhancing element particle prior to binding to the substrate. Other analytes will first bind with the substrate and then bind with an element particle. When a point-light source is shown through the sensor, a diffraction pattern is formed, indicating the presence of the analyte in the liquid.

10 The analytes that are contemplated as being detected using the present invention include, but are not limited to, bacteria; yeasts; fungi; viruses; rheumatoid factor; antibodies, including, but not limited to IgG, IgM, IgA and IgE antibodies; carcinoembryonic antigen; streptococcus Group A antigen; viral antigens; antigens associated with autoimmune disease; allergens; tumor antigens; streptococcus Group B antigen; HIV I or HIV II antigen; or host response (antibodies) to these and other viruses; 15 antigens specific to RSV or host response (antibodies) to the virus; an antigen; enzyme; hormone; polysaccharide; protein; lipid; carbohydrate; drug or nucleic acid; *Salmonella species*; *Candida species*, including, but not limited to *Candida albicans* and *Candida tropicalis*; *Salmonella species*; *Neisseria meningitidis* groups A, B, C, Y and W sub 135, *Streptococcus pneumoniae*, *E. coli K1*, *Haemophilus influenza* type B; an antigen derived from microorganisms; a hapten, a drug of abuse; a therapeutic drug; an environmental agent; and antigens specific to Hepatitis. 25

30 In another embodiment of the present invention, nutrients for a specific class of microorganisms can be incorporated into the analyte-specific receptor layer. In this way, very low concentrations of microorganisms can be detected by first contacting the biosensor of the present invention with the nutrients incorporated therein and then incubating the biosensor under conditions appropriate for the growth of the bound microorganism. The microorganism is allowed to grow until there are enough organisms to form a diffraction pattern. Of 35 course, in some cases, the microorganism is present or can multiply enough to form a diffraction pattern without the presence of a nutrient on the patterned monolayer.

A part of the present invention is the analyte-specific receptor material that can be microprinted on the polymer film and will specifically bind to the analyte of interest. Thus, the receptor material is defined as one part of a specific binding pair and includes, but is not limited to, antigen/ antibody, enzyme/substrate, oligonucleotide/DNA, chelator/metal, enzyme/inhibitor, bacteria/receptor, virus/receptor, hormone/receptor, DNA/RNA, or RNA/RNA, oligonucleotide/RNA, and binding of these species to any other species, as well as the interaction of these species with inorganic species. Additionally, when a metalized polymer film is used, the analyte-specific receptor material can be microprinted on the metalized surface of the film.

The receptor material that is bound to the attachment layer is characterized by an ability to specifically bind the analyte or analytes of interest. The variety of materials that can be used as receptor material are limited only by the types of material which will combine selectively (with respect to any chosen sample) with the analyte. Subclasses of materials which can be included in the overall class of receptor materials includes toxins, antibodies, antigens, hormone receptors, parasites, cells, haptens, metabolites, allergens, nucleic acids, nuclear materials, autoantibodies, blood proteins, cellular debris, enzymes, tissue proteins, enzyme substrates, coenzymes, neuron transmitters, viruses, viral particles, microorganisms, proteins, polysaccharides, chelators, drugs, and any other member of a specific binding pair. This list only incorporates some of the many different materials that can be coated onto the attachment layer to produce a thin film assay system. Whatever the selected analyte of interest is, the receptor material is designed to bind with the analyte of interest. In the preferred embodiments, the biosensing device is configured and arranged to provide a pattern detectable by eye in response to transmission of a point light source when the analyte of interest is sandwiched between the receptor material and a diffraction enhancing element.

In many instances, a "blocker" may be necessary to prevent non-specific binding. The term "blocker" as used herein

means a reagent that adheres to the sensor surface so that it "blocks" or prevents non-analyte materials from binding to the surface (either in the patterned or un-patterned areas). The blocking step may be done as a post-treatment to a surface which has already been contact printed ("post-block"), and is the standard technique for filling in non-contact printed regions with another thiol. However, the inventors have discovered that a "pre-block" technique is preferred over the post-block technique. In the pre-block technique, the surface of the substrate is pre-treated with a non-thiol containing blocker and then contact printed. Not wishing to be bound to any theory, it is theorized that the contact printed material (usually sulfur containing) displaces the physisorbed blocker, thereby permitting the analyte-specific receptor material to be bound directly to the surface of the substrate. A subsequent post-block may also be performed, if desired. Blockers can include, but are not limited to, β -casein, albumins such as bovine serum albumin, pluronic or other surfactants, polyethylene glycol, polyvinyl alcohol, or sulfur derivatives of the above compounds, and any other blocking material known to those of ordinary skill in the art.

The matrix containing the analyte of interest may be an interstitial fluid, a solid, a gas, or a bodily fluid such as mucous, saliva, urine, fecal material, tissue, marrow, cerebral spinal fluid, serum, plasma, whole blood, sputum, buffered solutions, extracted solutions, semen, vaginal secretions, pericardial, gastric, peritoneal, pleural, a throat swab or other washes and the like. The analyte of interest may be an antigen, an antibody, an enzyme, a DNA fragment, an intact gene, a RNA fragment, a small molecule, a metal, a toxin, an environmental agent, a nucleic acid, a cytoplasm component, pili or flagella component, protein, polysaccharide, drug, or any other material. For example, receptor material for bacteria may specifically bind a surface membrane component, protein or lipid, a polysaccharide, a nucleic acid, or an enzyme. The analyte which is indicative of the bacteria may be a saccharide or polysaccharide, an enzyme, a nucleic acid, a membrane component, a ganglioside or an antibody produced by the host in response to the bacteria. The

5 presence of the analyte may indicate an infectious disease (bacterial or viral), cancer, an allergy, or other medical disorder or condition. The presence of the analyte may be an indication of water or food contamination or other harmful materials. The analyte may indicate drug abuse or may monitor levels of therapeutic agents.

10 One of the most commonly encountered assay protocols for which this technology can be utilized is an immunoassay. However, the general considerations apply to nucleic acid probes, enzyme/substrate, and other ligand/receptor assay formats. For immunoassays, an antibody may serve as the receptor material and/or it may be the analyte of interest. The receptor material, for example an antibody or an antigen, must form a stable, reactive layer on the attachment layer of the test device. If an antibody is the receptor material, the antibody must be specific to the antigen of interest; and the antibody (receptor material) must bind the antigen (analyte) with sufficient avidity that the antigen is retained at the test surface. In some cases, the analyte may not simply bind the receptor material, but may cause a detectable modification of the receptor material to occur. This interaction could cause an increase in mass at the test surface or a decrease in the amount of receptor material on the test surface. An example of the latter is the interaction of a degradative enzyme or material with a specific, immobilized substrate. In this case, one would see a diffraction pattern before interaction with the analyte of interest, but the diffraction pattern would disappear if the analyte were present. The specific mechanism through which binding, hybridization, or interaction of the analyte with the receptor material occurs is not important to this invention, but may impact the reaction conditions used in the final assay protocol.

30 In general, the receptor material may be passively applied to the substrate layer. If required, the free functional groups introduced onto the test surface by the attachment layer may be used for covalent attachment of receptor material to the test surface.

5 A wide range of techniques can be used to apply the receptor material to the substrate layer. Test surfaces may be coated with receptor material by application of solution in discrete arrays or patterns; spraying, ink jet, contact printing or other imprinting methods; or printing a blocker material in a pattern followed by total immersion or spin coating with the receptor material. The technique selected should minimize the amount of receptor material required for coating a large number of test surfaces and maintain the stability/functionality of receptor material during application. The technique must also apply or adhere the receptor material to the attachment layer in a very uniform and controlled fashion.

10 The biosensing device of the present invention utilizes methods of contact printing of patterned, analyte-specific receptor layers on polymer or metalized polymer films, desirably transparent or semi-transparent, the compositions produced thereby, and the use of these compositions. Patterned analyte-specific receptor layers allow for the controlled attachment (or binding) placement of the analyte receptor. The term "patterned analyte-specific receptor layers thereon" as used herein means the analyte-specific receptor layers in any pattern on the polymer or metalized polymer films, including a solid pattern.

20 When the film with the patterned analyte-specific receptor layers thereon is exposed to an analyte that is capable of reacting with the analyte-specific receptor layer, the film will produce optical diffraction patterns which differ depending on the reaction of the patterned analyte-specific receptor layer with the analyte of interest. The medium would contain the diffraction enhancing element particles. The medium may be a high surface tension fluid such as water. The light can be in the visible spectrum, and be either reflected from the film, or transmitted through it, and the analyte can be any compound reacting with the analyte-specific receptor layer.

25 In preferred embodiments, the method involves contacting the sensing device with a test sample containing the diffraction enhancing element particles and potentially containing the analyte. If the analyte is present in the sample, then when light is

transmitted through a metalized polymer film with the analyte-specific receptor layer, a visible diffraction image is formed.

5 The medium in which the analyte may reside can be solid, gel-like, liquid or gas. For purposes of detecting an analyte in a body fluid, the fluid is selected from, but not limited to, urine, serum, plasma, spinal fluid, sputum, whole blood, saliva, urogenital secretions, fecal extracts, pericardial, gastric, peritoneal, pleural washes, vaginal secretions, or a throat swab. The most common gas that is contemplated as being used with the biosensing device of the present invention is air

10 In one embodiment, the present invention is contemplated in a dipstick form in which a micro-contact printed metalized film is mounted at the end of the dipstick. In use, the dipstick is dipped into the liquid in which the suspected analyte may be present. The liquid would also contain the diffraction enhancing element particles. The dipstick is allowed to remain for several minutes. The dipstick is then removed and then, either a light is projected through the metalized film or the film is observed with a light behind the film. If a diffraction image is observed, then the analyte is present in the liquid.

15 In another embodiment of the present invention, a multiple analyte test is constructed on the same support. As shown in Figure 1, a strip 10 is provided with several micro-contact printed films 20, 25, 30 and 35, each film having a pattern 40 printed thereon. Each of the micro-contact printed films 15, 20, 25, and 30 have a different receptor material that is specific for different analytes. It can be seen that the present invention can be formatted in any array with a variety of micro-contact printed films thereby allowing the user of the biosensor device of the present invention to detect the presence of multiple analytes in a medium using a single test.

20 There are many possible supports for the analyte-specific receptor layers. Simple physisorption can occur on many materials, such as polystyrene glass, nylon, or others well known to those of ordinary skill in the art. Preferred embodiments of immobilizing the analyte-specific receptor layers have involved covalent attachment, such as that possible between thiol or

disulfide-containing compounds and gold. Typically, a gold coating, 5 to 2000 nm thick, is supported on a Si/SiO₂ wafer, glass, or a polymer film. Optionally, titanium can be used to serve as an adhesion promoter between gold and the support.

5 The analyte-specific receptor attaches to the gold surface during contact printing or immersion from a solution. Preferably, the support comprises a gold coating on a MYLAR[®] film.

Figure 2 outlines the procedure used for microcontact printing. An elastomeric stamp is used to transfer analyte-specific receptor "ink" to a gold surface by contact; if the stamp is patterned, a patterned analyte-specific receptor layer forms.

10 The stamp is fabricated by casting polydimethylsiloxane (PDMS) on a master having the inverse of the desired pattern. Masters are prepared using standard photolithographic techniques, or constructed from existing materials having microscale surface features.

15

In a preferred embodiment of a typical experimental procedure, a photolithographically produced master is placed in a glass or plastic Petri dish, and a 10:1 ratio (w:w) mixture of SYLGARD[®] silicone elastomer 184 and SYLGARD[®] silicone elastomer 184 curing agent (Dow Corning Corporation) is poured over it. The elastomer is allowed to sit for approximately 30 minutes at room temperature and reduced pressure to degas, then cured for at least 4 hours at 60°C, and gently peeled from the master.

20

25 "Inking" of the elastomeric stamp is accomplished by exposing the stamp to a 0.1 to 10 μ M aqueous solution of disulfide-derivatized antibody typically by placing the stamp face down in the solution for 10 seconds to 10 minutes. The stamp is allowed to dry, either under ambient conditions, or typically by exposure to a stream of air or nitrogen gas. Following inking, the stamp is applied to a gold surface. Light pressure is used to ensure complete contact between the stamp and the surface.

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35 After 1 second to 5 minutes, the stamp is then gently peeled from the surface. Following removal of the stamp, the surface is rinsed and dried. Alternatively, further derivatization of unstamped areas can be accomplished, either by using a second stamp or by exposing the entire surface with a different reagent.

Subsequently, exposure to a protein-blocking agent, such as BSA or β -casein, or any other agent well known in the art, can also be done.

5 The elastomeric character of the stamp is important to the success of the process. Polydimethylsiloxane (PDMS), when cured, is sufficiently elastomeric to allow good conformal contact of the stamp and the surface, even for surfaces with significant relief; this contact is essential for efficient contact transfer of the receptor to a gold film. The elastomeric properties of PDMS are also important when the stamp is removed from the master: if
10 the stamp were rigid (as is the master) it would be difficult to separate the stamp and master after curing without damaging one of the two substrates. PDMS is also sufficiently rigid to retain its shape, even for features with sub-micron dimension. The stamp is durable in that the same stamp can be used over
15 200 times over a period of a year without significant degradation in performance. Using a printing roll for the stamp could allow for a continuous printing operation. Alternatively, ink-jet printing of the desired pattern could also be done if capable of producing the feature sizes needed for diffraction, for example \leq
20 100 μm .

A more detailed description of the methods and compositions of the present invention follows. All publications cited herein are incorporated by reference in their entirety.

25 Any plastic film is suitable for the present invention. Preferably, the plastic film is also capable of having a metal coating deposited thereon. These include, but are not limited to polymers such as: polyethylene-terephthalate (e.g., MYLAR®), acrylonitrile-butadiene-styrene, acrylonitrile-methyl acrylate
30 copolymer, cellophane, cellulosic polymers such as ethyl cellulose, cellulose acetate, cellulose acetate butyrate, cellulose propionate, cellulose triacetate, cellulose triacetate, polyethylene, polyethylene - vinyl acetate copolymers, ionomers (ethylene polymers) polyethylene-nylon copolymers, polypropylene,
35 methyl pentene polymers, polyvinyl fluoride, and aromatic polysulfones. Preferably, the plastic film has an optical transparency of greater than 80%. Other suitable plastics and

suppliers may be found, for example, in reference works such as the *Modern Plastics Encyclopedia* (McGraw-Hill Publishing Co., New York 1923-1996).

5 In one embodiment of the invention, the polymer film has a metal coating thereon and has an optical transparency of between approximately 5% and 95%. A more desired optical transparency for the plastic film used in the present invention is between approximately 20% and 80%. In a desired embodiment of the present invention, the polymer film has at least an
10 approximately 80% optical transparency, and the thickness of the metal coating is such as to maintain an optical transparency greater than about 60%, so that diffraction images can be produced by transmitted light. This corresponds to a metal coating thickness of about 10 nm. However, in other
15 embodiments of the invention, the gold thickness may be between approximately 1 nm and 1000 nm; for example, thicker gold coatings (>20 nm) would still be suitable for producing diffraction images by reflected light.

20 The preferred metal for deposition on the film is gold. However, silver, aluminum, chromium, copper, iron, zirconium, platinum and nickel, as well as oxides of these metals, may be used.

25 In principle, any surface with corrugations of appropriate size could be used as masters. The process of microcontact printing starts with an appropriate relief structure, from which an elastomeric stamp is cast. This 'master' template may be generated photolithographically, or by other procedures, such as commercially available diffraction gratings. In one embodiment, the stamp may be made from polydimethylsiloxane.

30 The stamp may be applied in air, or under a fluid capable of preventing excess diffusion of the receptor material. For large-scale or continuous printing processes, it is most desirable to print in air.

35 In one embodiment of the present invention, the pattern is formed on the metalized plastic polymer with the analyte-specific receptor layer. After the stamping process, the metalized areas

on the plastic may optionally be blocked, for example, with a protein-repelling agent such as β -casein.

5 This invention is further illustrated by the following examples, which are not to be construed in any way as imposing limitations upon the scope thereof. On the contrary, it is to be clearly understood that resort may be had to various other embodiments, modifications, and equivalents thereof, which, after reading the description herein, may suggest themselves to those skilled in the art without departing from the spirit of the present invention.

10

EXAMPLES

15 EXAMPLE 1

Antibody-conjugated polystyrene particles were produced by carbodiimide coupling with ethyldimethylaminodicarbodiimide (EDAC, bottle #3 of Polysciences kit, Catalog # 19539). For this example, 0.125 mL of a 10% suspension of 0.5 micron diameter blue carboxylated particles (Bangs Laboratories, Fishers, Indiana, Cat #D0005070CB) were activated with an aqueous solution of the EDAC for 1-4 hours, rinsed, then exposed to 300 micrograms of a monoclonal antibody to luteinizing hormone, alpha subunit, (Fitzgerald Industries, Concord, Massachusetts, Cat# 10-L10, Clone # M94136). The particles were again rinsed, blocked with bovine serum albumin, and stored at 2.5% concentration in phosphate buffered saline.

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Next, a gold/MYLAR® film was pre-treated (or blocked) with a 5 mg/mL beta casein solution for 10 minutes, then thoroughly rinsed and dried under an air stream. A PDMS stamp of 10-micron circles was coated with thiolated antibody by placing the stamp face down in a 0.5 mg/mL thiolated antibody solution and soaking for 10 minutes. A strong air stream was used to thoroughly dry the surface of the stamp. The coated stamp was placed in contact with the gold/MYLAR® film for 5

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minutes, then removed. The resulting printed gold/MYLAR® film was rinsed in distilled water, and dried.

5 A 10 mM stock solution of the Sulfo-LC-SPDP is prepared by dissolving 1.3 mg Sulfo-LC-SPDP into 2.07 ml de-ionized water. The conjugation reaction is carried out in phosphate buffered saline (PBS) containing 20 mM sodium phosphate buffer, 150 mM NaCl, 1mM EDTA and 0.02% sodium azide at pH 7.5. One milligram of lyophilized antibody is dissolved in 450 ml PBS, and 50 ml of Sulfo-LC-SPDP stock solution is added to the antibody solution. The mixture is allowed to react at room temperature for 60 minutes. The sample is applied to a 5 ml desalting polyacrylamide column previously equilibrated with 5 bed volumes (25 ml) of PBS. Fractions are eluted using PBS as the elution buffer, and the protein in the fractions is monitored using a COOMASSIE® Protein Assay (Pierce Chemical Co). Typically, 50 ul of the COOMASSIE® reagent is mixed with 50 ul of each fraction in a micro-titer plate. The COOMASSIE® Blue substrate reacts with the protein, producing a blue color, the intensity of which is dependent upon the amount of protein present in the fraction. The fractions which produce the most intense blue color are those containing the majority of the protein eluted. These fractions are pooled together to produce the disulfide form of the final derivatized product. This is typically the form used for contact printing.

25 Optionally, the disulfide-pyridyl group present on the disulfide form of the thiolated binder can be reduced to a thiol group in a reduction reaction. Instead of desalting on a column equilibrated with PBS, the derivatized protein is desalted on a column equilibrated with an acetate buffer (100 mM sodium acetate buffer, 100 mM NaCl, pH 4.5). The acidic pH of this acetate buffer acts to protect any disulfide bonds present on the native protein from undesired reduction. In the reduction reaction, 12 mg of dithiothreitol (DTT) is dissolved in 500 ml acetate buffer and added to 1 ml of the SPDP derivatized protein. The reaction mixture is incubated for 30 minutes at room temperature, and desalted on a 5 ml desalting column

equilibrated with 5 bed volumes (25 ml) of acetate buffer. The protein content of the fractions eluted is again monitored by the COOMASSIE® Protein Assay as described above, and the fractions containing the greatest amount of protein are pooled.

5 Both the disulfide and reduced forms of the thiolated binders are stored in aqueous solution at 4° C until used for contact printing.

10 The sensors were then used to detect an analyte. The analyte solution was then mixed with microparticles (typically 50-70 microliters of analyte solution in 1% bovine serum albumin with 10-25 microliters of 1.5-2.5% particle suspension; preferably, there is a 50:25 ratio of analyte solution to particle suspension), and placed on top of a 1 cm square sensor sample. After 5 minutes, a nitrocellulose disk (5 or 8 micron pore size, Sigma #N3771 or N4146) with a small hole (e.g. 3/16") punched out of the center was placed on top of the sensor. The disk was used to wick away excess fluid and unbound microparticles. At this time, a point light source was transmitted through the sensor sample (using the small hole in the nitrocellulose). A diffraction image would be seen on the other side of the light beam in the presence of the target analyte.

20 As seen in Figure 4, SEM photomicrographs showed the patterned placement of the microparticles.

25 EXAMPLE 2

A PDMS stamp of an x,y array of 10-micron circles was "inked" with thiolated 30-mer oligonucleotide which is complementary to the target DNA strand ("30-mer"; base sequence of thiol spacer-5'-
30 CAATCCACGTCACGGACAGGGTGAGGAAGA-3' made by Genosys, Inc., The Woodlands, Texas) by placing the stamp face down with weight in oven-dried (50°C, vacuum) mixture of the 30-mer and ethyl acetate on glass. After 10 minutes, the inked stamp was removed. At the same time, a gold/MYLAR® film
35 was pre-heated on a 60°C hot plate for 5 minutes. Printing was done by placing the inked PDMS stamp on top of the gold-coated side of MYLAR® at 60°C; weight and heat were

5 maintained during the 5 minute contact time. At this point, the stamp was removed and the printed gold/MYLAR® film was washed with distilled water, and air-dried. The gold/MYLAR® film sample was then blocked with a 2.5 mg/mL beta casein solution (in phosphate buffered saline, pH 7.2) for 10 minutes, and rinsed with distilled water and air-dried.

10 These sensors were used to test for target DNA. Hybridization of the target DNA to the capture DNA patterned on the sensor surface took place as follows: A pre-heated analyte solution (60°C water bath, 2 minutes) containing a DNA strand of interest (a biotinylated 70-mer from Genosys with base sequence of biotin-5'-GGTAGACCGGAGAGCTGTGTCACCATGTGGGTCCCGGT TGTCTTCCTCACCTGTCCGTGACGTGGATTG-3') was added to a pre-heated sensor (60° C hot plate, 5 minutes) and then 75 microliters was added to an approximately 1 cm square sensor for an additional 10 minutes heating. After this time, the sensor sample was rinsed with water, and air-dried for subsequent testing with microparticles. One variation to this method is that the analyte solution, e.g., during a PCR amplification, and the microparticles are exposed to the sensor at the same time.

25 Next, Streptavidin-coated, 1 micron diameter particles from Bangs Laboratories (Catalog # CP01N) were added in 20-30 microliter amounts, concentration of 2.4×10^{11} particles per mL, to the sensor. The sensor and particles were heated on a 60 °C hot plate for 10 minutes (covered, while ensuring that complete evaporation did not take place), and then rinsed gently with distilled water. After this, a point light source was transmitted through the sensor sample. A diffraction image would be seen on the other side of the light beam in the presence of the DNA analyte.

30 SEM photomicrographs show the patterned placement of the microparticles.

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EXAMPLE 3

Antibody-conjugated polystyrene particles were produced by carbodiimide coupling with ethyldimethylaminodicarbodiimide ("EDAC", bottle #3 of Polysciences kit, Catalog # 19539). For example, 0.125 mL of a 10% suspension of 0.3 micron diameter blue carboxylated particles (Bangs Laboratories, Cat #DC02/1836) were activated with an aqueous solution of the EDAC for 1-4 hours, rinsed, then exposed to 300 micrograms of a polyclonal antibody to IgE (Fitzgerald Industries, Cat#20-IR77). The particles were again rinsed, blocked with bovine serum albumin, and stored at 1.7% concentration in phosphate buffered saline.

Next, a gold/MYLAR® film was pre-treated (or blocked) with a 5 mg/mL beta casein solution for 10 minutes, then thoroughly rinsed and dried under an air stream. A PDMS stamp of an x,y array of 10-micron diameter circles was coated with thiolated antibody (antibody was initially Fitzgerald Catalog #10-I10 then derivatized or "thiolated" using Sulfo-LC-SPDP by Pierce) by placing the stamp face down in a 0.5 mg/mL thiolated antibody solution and soaking for 10 minutes. A strong air stream was used to thoroughly dry the surface of the stamp. The coated stamp was placed in contact with the gold/MYLAR® film for 5 minutes, then removed. The resulting printed gold/MYLAR® film was rinsed in distilled water, and dried.

The analyte solution was then mixed with microparticles (typically 50-70 microliters of analyte solution in 1% bovine serum albumin with 10-25 microliters of 1.5-2.5% particle suspension; preferably, there is a 50:25 ratio of analyte solution to particle suspension), and placed on top of a 1 cm square sensor sample. After 5-10 minutes, a nitrocellulose disk (5 or 8 micron pore size, Sigma #N3771 or N4146) with a small (e.g., 3/16" diameter) hole punched out of the center is placed on top of the sensor. The disk was used to wick away excess fluid and unbound microparticles. At this time, a point light source was transmitted through the sensor sample by taking advantage of the small hole in the nitrocellulose. A high order diffraction

image was seen on the other side of the light beam, signifying the presence of the analyte.

EXAMPLE 4

5 A gold/MYLAR® film was pre-treated (or blocked) with a 5 mg/mL beta casein solution in phosphate buffered saline (pH ~ 7.2) for 10 minutes, then thoroughly rinsed and dried under an air stream. A PDMS stamp of 10-micron circles was coated with thiolated antibody (e.g., rabbit anti-Candida albicans, Cat # 20-CR04 from Fitzgerald Industries, Inc.) by placing the stamp face down in a 0.5 mg/mL thiolated antibody solution and soaking for 10 minutes. A strong air stream was used to thoroughly dry the surface of the stamp. The coated stamp was placed in contact with the gold/MYLAR® film for 2 minutes, then removed. The resulting printed gold/MYLAR® film was rinsed in distilled water, and dried.

10 The sensor sample was exposed to a 10% dilution in phosphate buffered saline, pH 7.2 of 40 nm gold particles coated with goat anti-rabbit IgG (gold conjugate was from Polysciences, Catalog # 22705). After one hour, the samples were thoroughly rinsed with distilled water and dried under a nitrogen or air stream. At this point, the samples do not diffract a HeNe laser beam.

20 The samples were then exposed to silver enhancing reagents from BBI (either BBI International's kit # SEKL 15 (Batch #2575) or large kit # SEKB250 (Batch #2484) were used). A 1:1 v/v ratio of the enhancer and initiator reagents in the kit were pre-mixed and then immediately placed on top of the gold-particle coated samples. After 10-20 minutes exposure (preferably, 10 minutes), the samples were rinsed with water, dried, and examined. At this point, the samples diffracted light (either a laser beam or a point white light source) most likely due to the larger size of the silver nucleated around the gold nanoparticles.

35

EXAMPLE 5

5 Samples prepared as per Examples 1 or 4 could also be developed into a diffraction image by exposing it to an enzyme-conjugate secondary antibody in the presence of the analyte, such that if the analyte is present the secondary antibody would bind and cause subsequent precipitate development with a precipitating substrate specific to the enzyme.

10 A gold/MYLAR® film was pre-treated (or blocked) with a 5 mg/mL beta casein solution in phosphate buffered saline (pH ~ 7.2) for 10 minutes, then thoroughly rinsed and dried under an air stream. A PDMS stamp of 10-micron circles was coated with thiolated antibody (e.g., mouse anti-luteinizing hormone beta, Cat # 10-L15 from Fitzgerald Industries, Inc.) by placing the stamp face down in a ~ 0.3 mg/mL thiolated antibody
15 solution and soaking for 10 minutes. A strong air stream was used to thoroughly dry the surface of the stamp. The coated stamp was placed in contact with the gold/MYLAR film for 5 minutes, then removed. The resulting printed gold/MYLAR® film was rinsed in distilled water, and dried.

20 The sensor sample was exposed to an analyte solution of luteinizing hormone (Cat # 30-AL15 from Fitzgerald Industries, Inc.) in 1% bovine serum albumin, phosphate buffered saline. Concentration of antigen was varied from 0.1 to 1000 ng / mL. After one hour at room temperature, the sample was rinsed with
25 0.02% TWEEN 20 solution, then distilled water. A subsequent exposure to a secondary antibody (Fitzgerald Catalog # 61-L05 diluted 1:100 in distilled water) for one hour was done, followed by rinsing as above. A TMB membrane enhancer solution (e.g., a 10:1 v/v mixture of Kirkegaard and Perry Laboratories' reagents Cat #50-76-18 and Cat#50-77-01) was placed on the
30 sample for 10 minutes to cause development of a blue precipitate in the circles or features. This precipitate caused a diffraction image to form upon irradiation with a point light source.

We claim:

1. A method of detecting an analyte in a medium comprising:
5 adding a diffraction enhancing element to the medium suspected of containing the analyte, wherein the diffraction enhancing element has a receptor material thereon that is specific for the analyte;
contacting the medium with a sensing device, the sensing
10 device comprising:
a polymer film; and
an analyte-specific receptor layer printed in a pattern onto the polymer film wherein the analyte-specific receptor layer has a receptor material thereon that is specific for the analyte;
15 transmitting a light through the polymer film; and
detecting presence of the analyte by detecting a pattern formed by diffraction of the transmitted light.
2. The method of Claim 1, wherein the analyte-specific
20 receptor layer is printed in a pattern such that when the sensing device binds an analyte, the sensing device diffracts transmitted light to form a diffraction pattern.
3. The method of Claim 1, wherein the diffraction pattern is
25 visible to an unaided eye.
4. The method of Claim 1, further comprising a metal
coating on the polymer film and wherein the analyte-specific
30 receptor layer is printed onto the metal coating.
5. The method of Claim 4, wherein the metal is selected from gold, silver, chromium, nickel, platinum, aluminum, iron, copper, gold oxide, chromium oxide or zirconium.
- 35 6. The method of Claim 5, wherein the metal is gold.

7. The method of Claim 6, wherein the gold coating is between approximately 1 nanometer and 1000 nanometers in thickness.
- 5 8. The method of Claim 1, wherein the polymer film is selected from polyethylene-terephthalate, acrylonitrile-butadiene-styrene, acrylonitrile-methyl acrylate copolymer, cellophane, cellulosic polymers such as ethyl cellulose, cellulose acetate, cellulose acetate butyrate, cellulose propionate, cellulose triacetate, polyethylene, polyethylene - vinyl acetate copolymers, ionomers (ethylene polymers) polyethylene-nylon copolymers, polypropylene, methyl pentene polymers, polyvinyl fluoride, or aromatic polysulfones.
- 10
- 15 9. The method of Claim 8, wherein the polymer film is polyethylene-terephthalate.
- 20 10. The method of Claim 1, wherein the polymer film is optically transparent.
- 25 11. The method of Claim 10, wherein the polymer film has an optical transparency between 5% and 95%.
- 30 12. The method of Claim 10, wherein the polymer film has an optical transparency between approximately 20% and 80%.
13. The method of Claim 1, wherein there are two or more analyte-specific receptor layers with each layer having different chemical properties.

14. The method of Claim 1, wherein the analyte is selected from bacteria, yeast, fungus, virus, rheumatoid factor, IgG, IgM, IgA and IgE antibodies, carcinoembryonic antigen, streptococcus Group A antigen, viral antigens, antigens associated with autoimmune disease, allergens, tumor antigens, streptococcus Group B antigen, HIV I or HIV II antigen, antibodies viruses, antigens specific to RSV,, an antibody, antigen, enzyme, hormone, polysaccharide, protein, lipid, carbohydrate, drug or nucleic acid, *Neisseria meningitides* groups A, B, C, Y and W sub 135, *Streptococcus pneumoniae*, *E. coli* K1, *Haemophilus influenza* type B, an antigen derived from microorganisms, a hapten, a drug of abuse, a therapeutic drug, an environmental agent, or antigens specific to Hepatitis.
15. The method of Claim 14, wherein the analyte is bacteria, yeast, fungus or virus.
16. The method of Claim 1, wherein the receptor material is selected from antigens, antibodies, oligonucleotides, chelators, enzymes, bacteria, yeasts, fungi, viruses, bacterial pili, bacterial flagellar materials, nucleic acids, polysaccharides, lipids, proteins, carbohydrates, metals, hormones or receptors for said materials.
17. The method of Claim 1, wherein the diffraction enhancing element is selected from glass, cellulose, synthetic polymers or plastics, latex, polystyrene, polycarbonate, bacterial or fungal cells.
18. The method of Claim 1, wherein the diffraction enhancing element is polystyrene latex microspheres.
19. The method of Claim 1, further comprising the step of applying a blocking material to the non-printed areas of the polymer film.

20. The method of Claim 19, wherein the blocking material is selected from β -casein, an albumin, a surfactant, polyethylene glycol, polyvinyl alcohol, or sulfur derivatives thereof.
- 5 21. The method of Claim 1, wherein the sensing device further comprises a layer of blocking material on the polymer film through which the analyte-specific receptor material is printed.
- 10 22. The method of Claim 21, wherein the blocking material is selected from β -casein, an albumin, a surfactant, polyethylene glycol, polyvinyl alcohol, or sulfur derivatives thereof.
- 15 23. A method of detecting an analyte in a medium comprising:
adding a diffraction enhancing element to the medium suspected of containing the analyte, wherein the diffraction enhancing element has a receptor material thereon that is specific for the analyte;
20 contacting the medium with a sensing device, the sensing device comprising:
a polymer film coated with metal; and
an analyte-specific receptor layer printed in a pattern onto the metal-coated polymer film wherein the analyte-specific
25 receptor layer has a receptor material thereon that is specific for the analyte;
reflecting a light source off a surface of the metal-coated polymer film; and
detecting presence of the analyte by detecting a pattern
30 formed by diffraction of the reflected light.
- 35 24. The method Claim 23, wherein the analyte-specific receptor layer is printed in a pattern such that when the sensing device binds an analyte, the sensing device diffracts reflected light to form a diffraction pattern.

25. The method of Claim 23, wherein the diffraction pattern is visible to an unaided eye.
- 5 26. The method of Claim 23, wherein the metal is selected from gold, silver, chromium, nickel, platinum, aluminum, iron, copper, gold oxide, chromium oxide or zirconium.
27. The method of Claim 26, wherein the metal is gold.
- 10 28. The method of Claim 27, wherein the gold coating is between approximately 1 nanometer and 1000 nanometers in thickness.
- 15 29. The method of Claim 23, wherein the polymer film is selected from polyethylene-terephthalate, acrylonitrile-butadiene-styrene, acrylonitrile-methyl acrylate copolymer, cellophane, cellulosic polymers such as ethyl cellulose, cellulose acetate, cellulose acetate butyrate, cellulose propionate, cellulose triacetate, polyethylene, polyethylene-vinyl acetate copolymers, ionomers (ethylene polymers) polyethylene-nylon copolymers, 20 polypropylene, methyl pentene polymers, polyvinyl fluoride, or aromatic polysulfones.
- 25 30. The method of Claim 29, wherein the polymer film is polyethylene-terephthalate.
- 30 31. The method of Claim 23, wherein there are two or more analyte-specific receptor layers with each layer having different chemical properties.

32. The method of Claim 23, wherein the analyte is selected from bacteria, yeast, fungus, virus, rheumatoid factor, IgG, IgM, IgA and IgE antibodies, carcinoembryonic antigen, streptococcus Group A antigen, viral antigens, antigens associated with autoimmune disease, allergens, tumor antigens, streptococcus Group B antigen, HIV I or HIV II antigen, antibodies viruses, antigens specific to RSV,, an antibody, antigen, enzyme, hormone, polysaccharide, protein, lipid, carbohydrate, drug or nucleic acid, *Neisseria meningitides* groups A, B, C, Y and W sub 135, *Streptococcus pneumoniae*, *E. coli* K1, *Haemophilus influenza* type B, an antigen derived from microorganisms, a hapten, a drug of abuse, a therapeutic drug, an environmental agent, or antigens specific to Hepatitis.

33. The method of Claim 32, wherein the analyte is bacteria, yeast, fungus or virus.

34. The method of Claim 23, wherein the receptor material is selected from antigens, antibodies, oligonucleotides, chelators, enzymes, bacteria, yeasts, fungi, viruses, bacterial pili, bacterial flagellar materials, nucleic acids, polysaccharides, lipids, proteins, carbohydrates, metals, hormones or receptors for said materials.

35. The method of Claim 23, wherein the diffraction enhancing element is selected from glass, cellulose, synthetic polymers or plastics, latex, polystyrene, polycarbonate, bacterial or fungal cells.

36. The method of Claim 23, wherein the diffraction enhancing element is polystyrene latex microspheres.

37. The method of Claim 23, further comprising the step of applying a blocking material to the non-printed areas of the metal-coated polymer film.

38. The method of Claim 37, wherein the blocking material is selected from β -casein, an albumin, a surfactant, polyethylene glycol, polyvinyl alcohol, or sulfur derivatives thereof.
- 5 39. The method of Claim 23, wherein the sensing device further comprises a layer of blocking material on the metal-coated polymer film through which the analyte-specific receptor material is printed.
- 10 40. The method of Claim 39, wherein the blocking material is selected from β -casein, an albumin, a surfactant, polyethylene glycol, polyvinyl alcohol, or sulfur derivatives thereof.
- 15 41. A method of detecting an analyte in a medium comprising:
 adding a diffraction enhancing element to the medium suspected of containing the analyte, wherein the diffraction enhancing element has a receptor material thereon that is specific for the analyte;
20 contacting the medium with a sensing device, the sensing device comprising:
 a polymer film coated with metal; and
 an analyte-specific receptor layer printed in a pattern onto the metal-coated polymer film wherein the analyte-specific receptor
25 layer has a receptor material thereon that is specific for the analyte;
 transmitting a light through the polymer film; and
 detecting presence of the analyte by detecting a pattern formed by diffraction of the transmitted light.
- 30

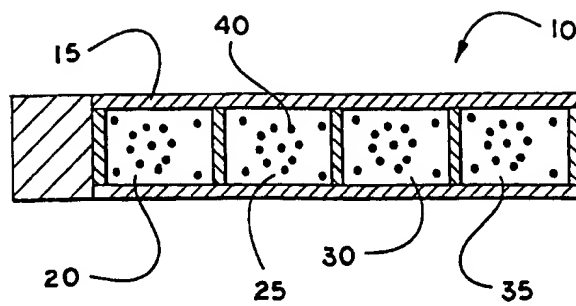


FIG. 1

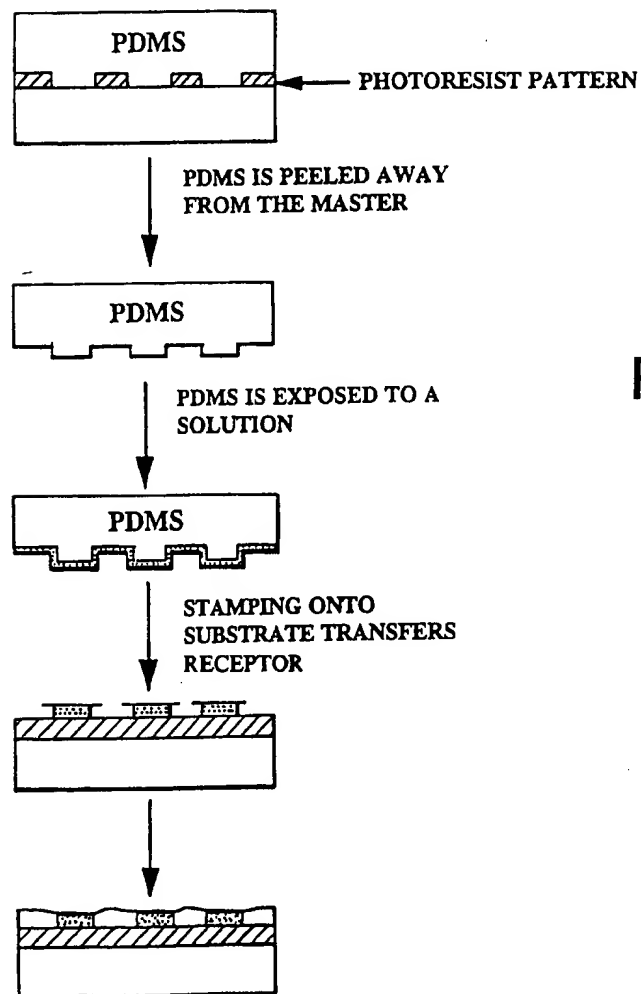


FIG. 2

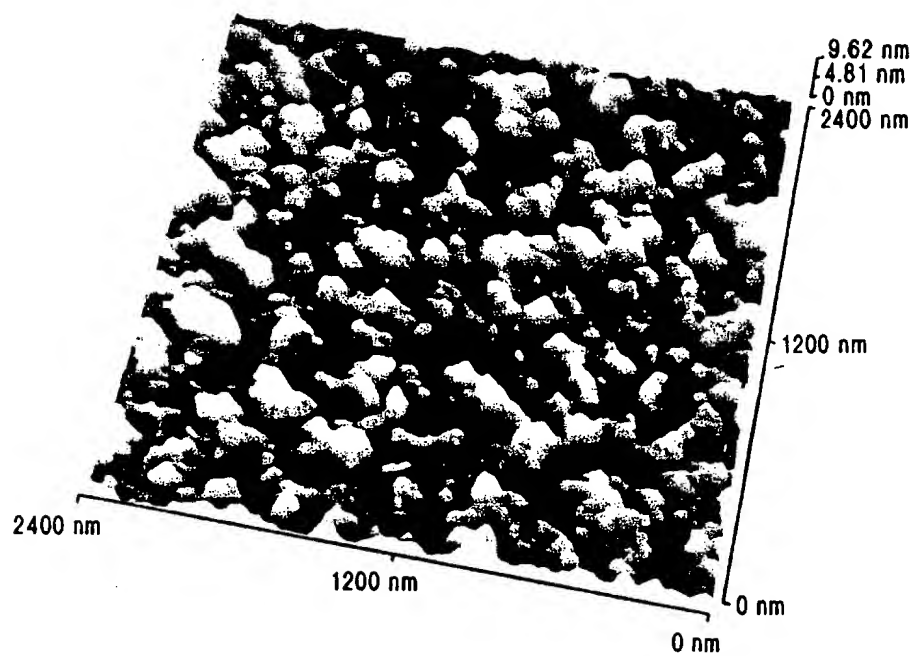


FIG. 3

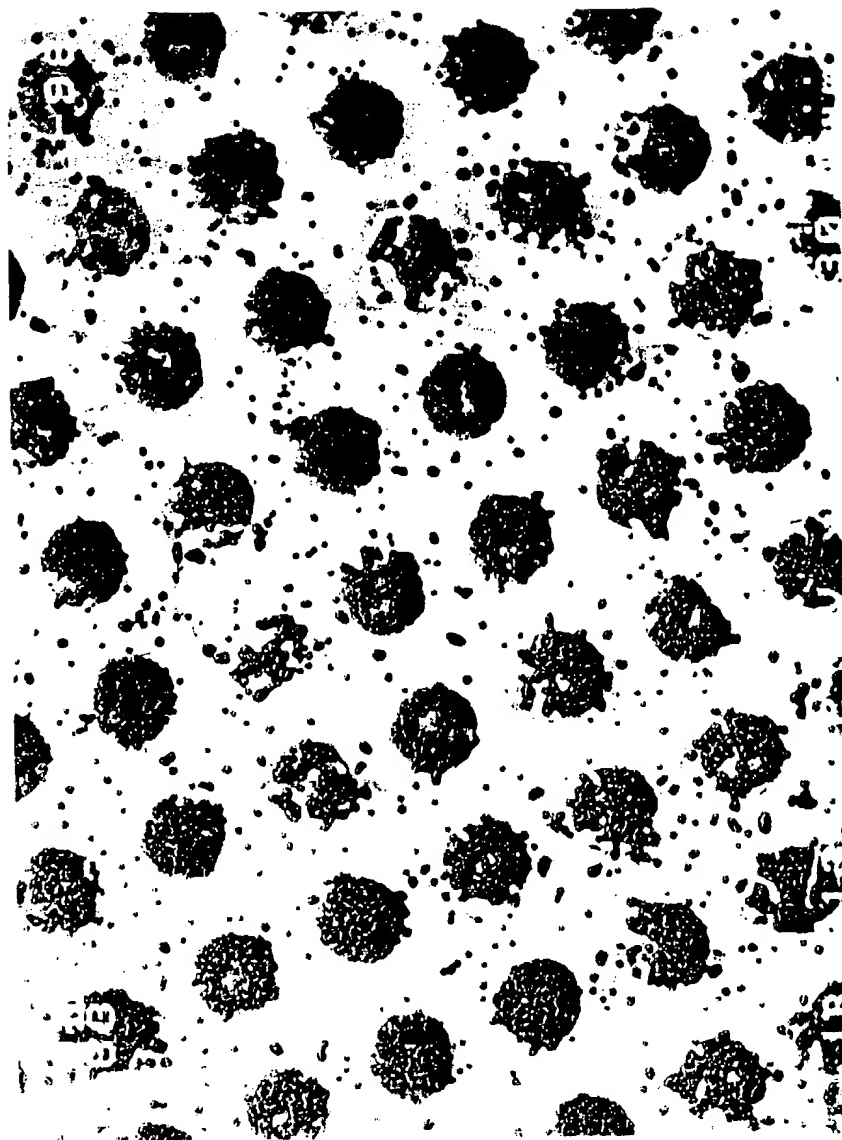


FIG. 4

INTERNATIONAL SEARCH REPORT

International Application No.
PCT/AT00/00167

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 H02K16/00 H02K16/02

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 H02K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 769 403 A (TOYOTA MOTOR CO LTD) 23 April 1997 (1997-04-23) paragraph '0006! column 8, line 1 - line 41; figures 1,2	1-3,6-8, 11
A	EP 0 725 474 A (NIPPON DENSO CO) 7 August 1996 (1996-08-07) column 4, line 12 - line 20 column 4, line 44 - line 51 column 5, line 15 - line 32 column 6, line 30 - line 33; figure 1	1-3,6-9
A	EP 0 800 951 A (TOYOTA MOTOR CO LTD) 15 October 1997 (15-10-1997) Abstract, Claim 1 figure 1	1-3,6-8

☐ Further documents are cited in the continuation of box C.

☒ Patent family members are listed in annex.

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Date of the actual completion of the international search

9 November 2000

Date of mailing of the international search report

16/11/2000

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Information on patent family members

International application No

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